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NOUVELLE SÉRIE

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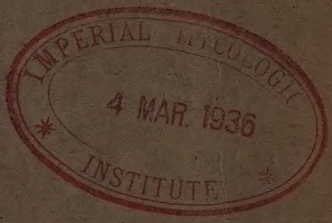

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MATHÉMATIQUES

SUR QUELQUES MÉTHODES PARTICULIÈRES DE PROLONGEMENT DE L'ESPACE DE HILBERT

Par. L. V. KANTOROVIČ (KANTOROVITCH)

(*Présenté par S. N. Bernstein, de l'Académie, le 5. IX. 1935*)

Dans ma précédente note* j'ai donné une méthode générale de prolongement de l'espace abstrait de Hilbert. En choisissant de façon appropriée la famille d'opérations B et l'ensemble $\mathfrak{B}$ nous obtenons divers procédés particuliers de prolongement de l'espase de Hilbert.

Dant cette note je considère trois réalisations de l'espace de Hilbert: 1) au moyen de suites de nombres dont la série des carrés est convergente, 2) au moyen de fonctions à carré sommables dans un intervalle fini, 3) au moyen de fonctions à carré sommables dans l'intervalle $<-\infty, +\infty>$.

Dans ces cas, en appliquant telle ou telle autre famille d'opérations B, tel ou tel ensemble $\mathfrak{B}$, j'obtiens des prolongements différents de tous les trois espaces. De plus, dans tous ces cas l'espace prolongé $\mathfrak{H}$ se laisse également réaliser de façon déterminée.

I. Soit H_1 l'espace des suites de nombres complexes:

$$H_1 = \{ x \}; \quad x = (x_1, x_2, \ldots),$$

$\sum |x_i|^2$ étant convergente. Le produit scalaire, si $x = (x_1, x_2, \ldots)$; $y = (y_1, y_2, \ldots)$ est défini comme $(x, y) = \sum_i x_i \bar{y}_i$.

Prenons pour l'ensemble $\mathfrak{B}$ l'ensemble de points x, dont toutes les coordonnées, à partir d'un certain x_n, sont égales à zéro, c.-à-d. que

$$\mathfrak{B} = \{ (x_1, x_2, \ldots, x_n, 0, 0, \ldots) \}.$$

A chaque suite de nombres positifs $\gamma_1, \gamma_2, \ldots$; $\gamma_i > 0$ correspond l'opération B définie dans $\mathfrak{B}$ comme

$$B(x_1, x_2, \ldots, x_n, 0, 0, \ldots) = (\gamma_1 x_1, \gamma_2 x_2, \ldots, \gamma_n x_n, 0, 0, \ldots).$$

Adoptons pour $B = \{ B \}$ la famille d'opérations B correspondant à toutes les suites $\gamma_1, \gamma_2, \ldots$ Cette famille B et l'ensemble $\mathfrak{B}$ satisfont à toutes les conditions 1)—7). En effet:

1. B est une opération additive définie dans $\mathfrak{B}$. Le domaine de ses valeurs dans $\mathfrak{B}$ est également situé dans $\mathfrak{B}$. L'ensemble $\mathfrak{B}$ est dense dans H_1.

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* C. R. Acad. Sci. URSS, IV, p. 123 (1935). Dans la suite je me sers des désignations et des résultats de cette note sans m'y référer davantage.

2. L'opération B est symétrique dans $\mathfrak{B}$. En effet, on a:

$$(x, Bx) = \sum_{i=1}^{n} x_i \overline{\gamma_i x_i} = \sum_{i=1}^{n} \gamma_i |x_i|^2 = \sum_{i=1}^{n} \gamma_i x_i \bar{x}_i = (Bx, x).$$

3) L'opération B est positive définie:

$$(x, Bx) = \sum_{i=1}^{n} \gamma_i |x_i|^2 \geqslant 0,$$

l'égalité ayant lieu pour $x_i = 0$ $(i = 1, 2, \ldots)$ et dans ce cas seulement.

4) L'opération B^{-1} appartient à la famille B. En effet, cette opération correspond à la suite $\frac{1}{\gamma_1}, \frac{1}{\gamma_2}, \ldots$, et par conséquent entre dans B.

5) L'opération identique I appartient à la famille B. En effet, elle correspond à la suite $1, 1, \ldots$

6) Soit $B_1 \sim \gamma_1', \gamma_2', \ldots$; $B_2 \sim \gamma_1'', \gamma_2'', \ldots$; formons l'opération $B \sim \sim \gamma_1, \gamma_2, \ldots$, où $\gamma_i = \min(\gamma_i', \gamma_i'')$; alors on a:

$$\|x\|_B = \sqrt{\sum \gamma_i |x_i|^2} \leqslant \|x\|_{B_1}; \quad \|x\|_B \leqslant \|x\|_{B_2}; \quad x \in \mathfrak{B}.$$

7) Si $B \sim \gamma_1, \gamma_2, \ldots$ et $B' \sim \gamma_1', \gamma_2', \ldots$, alors l'opération BB' correspond à la suite $\gamma_1\gamma_1', \gamma_2\gamma_2', \ldots$ et par conséquent BB' appartient à B.

Si la suite d'éléments $x^{(\nu)} = (x_1^{(\nu)}, x_2^{(\nu)}, \ldots)$ est convergente B en soi, alors on aura $\sum \gamma_i |x_i^{(p)} - x_i^{(q)}|^2 \to 0$, où $p, q \to \infty$. Or, dans ces conditions, chaque coordonnée a une limite $\lim\limits_{\nu \to \infty} x_i^{(\nu)} = g_i$, la somme $\sum \gamma_i |g_i|^2$ étant convergente. Cela étant, à une telle suite $(g_1, g_2, \ldots)$ ne répondent que des suites équivalentes $\{x^{(\nu)}\}$. Ainsi dons, nous pouvons réaliser les éléments idéaux ajoutés par des suites $g = (g_1, g_2, \ldots)$ à somme $\sum \gamma_i |g_i|^2$ convergente. L'ensemble de pareilles suites formera $\mathfrak{G}_B$. L'espace prolongé total $\mathfrak{H}_1$ sera composé de toutes les suites possibles $h = (h_1, h_2, \ldots)$.

Le produit scalaire de deux éléments $g = (g_1, g_2, \ldots)$ et $h = (h_1, h_2, \ldots)$ est défini, si la somme $\sum |g_i| \cdot |h_i|$ converge et alors on a $(g, h) = \sum_{i=1}^{\infty} g_i h_i$.

En effet, admettant

$$\gamma_i = \begin{cases} \frac{|h_i|}{|g_i|}, & \text{si } h_i \neq 0; \quad g_i \neq 0; \\ \frac{1}{i^2 |g_i|^2}, & \text{si } g_i \neq 0; \quad h_i = 0; \\ i^2 |h_i|^2, & \text{si } g_i = 0; \quad h_i \neq 0; \\ 1, & \text{si } g_i = 0; \quad h_i = 0; \end{cases}$$

nous voyons que, si $B \sim \gamma_1, \gamma_2, \ldots$, alors on a $g \in \mathfrak{G}_B$ et $h \in \mathfrak{G}_{B^{-1}}$, donc (g, h) est défini.

Tous les théorèmes généraux indiqués dans le précédent article sont valables également pour cet espace.

Envisageons, par exemple, l'opération A définie dans $\mathfrak{B}$ par l'égalité

$$A(x_1, x_2, \ldots, x_n, 0, 0, \ldots) = (k_1 x_1, k_2 x_2, \ldots, k_n x_n, 0, 0, \ldots),$$

où $k_1, k_2, \ldots$ sont des nombres complexes quelconques. Cette opération est bornée B, si $B \sim \gamma_1, \gamma_2, \ldots$ où $\gamma_i = |k_i|$. C'est pourquoi elle s'étend à tout l'espace $\mathfrak{H}_1$ et l'on a évidemment:

$$A(h_1, h_2, \ldots) = (k_1 h_1, k_2 h_2, \ldots).$$

L'opération inverse A^{-1} sera bornée B^{-1} et également définie dans tout l'espace $\mathfrak{H}_1$ (pourvu que $k_i \neq 0$).

Remarque 1. Notons que l'espace ici défini $\mathfrak{H}_1$, est doué d'une compacticité singulière. Tout d'abord, s'il est donné une suite d'éléments de $\mathfrak{H}_1 : h^\nu = (h_1^\nu, h_2^\nu, \ldots)$ avec des limites existantes $h_i = \lim\limits_{\nu\to\infty} h_i^\nu$, il est possible de trouver une opération $B \sim \gamma_1, \gamma_2, \ldots$ telle que les points h^ν seront convergents B vers h. Ensuite, si au sujet de la suite h^ν il est seulement connu que les h_i^ν sont bornés: $|h_i^\nu| < M_i (\nu = 1, 2, \ldots)$, alors il est possible d'en choisir une suite partielle h^{ν_k} qui convergera B vers un certain élément h faisant partie de l'espace $\mathfrak{H}_1$.

Remarque 2. Citons un exemple de prolongement d'une opération non bornée. Considérons seulement pour la facilité du raisonnement des suites bilatérales $x = (\ldots x_1, x_0, x_1, x_2, \ldots)$. Soit $c = (\ldots, c_1, c_0, c_1, \ldots)$ un certain élément de H_1. Il lui correspond l'opération $Cx = u$; $u = (u_1, u_2, \ldots)$, où $u_p = \sum\limits_{\nu=-\infty}^{+\infty} C_{p-\nu} x_\nu$ transformant $\mathfrak{B}$ en une partie de H_1. La somme ne contenant qu'un nombre fini de membres, si x fait partie de $\mathfrak{B}$, Cx est défini dans $\mathfrak{B}$. Il est alors aisé d'établir que, si l'espace $\mathfrak{G}_B$, $B \sim \gamma_1, \gamma_2, \ldots$ est tel que toutes les suites $\{c_{p-\nu}\}$ lui appartiennent, c.-à-d. que la somme $\sum\limits_\nu \gamma_\nu |c_{p-\nu}|^2$ converge, l'opération Cx est définie dans l'espace $\mathfrak{G}_{B^{-1}}$, c.-à-d. pour les éléments g à $\sum\limits_\nu \frac{|g_\nu|^2}{\gamma_\nu}$ convergente. La valeur de l'opération $u = Cg$ appartient à H_1 lorsque la série $\sum\limits_p \left(\sum\limits_\nu \gamma_\nu |c_{p-\nu}|^2\right)$ est convergente.

Remarque 3. Notons que l'espace prolongé de Hilbert ici traité peut être également appliqué lors de sa réalisation fonctionelle. Notamment, si l'on introduit des coordonnées dans l'espace de fonctions à carré sommables à l'aide d'un certain système orthogonal complet. Toutefois, le prolongement de l'espace ainsi obtenu dépend essentiellement du système orthogonal placé à sa base.

Remarque 4. En particulier, il est permis de prendre pour système fondamental (rem. 3) les polynomes de Legendre. Alors, l'ensemble $\mathfrak{B}$ sera l'ensemble de tous les polynomes ordinaires. Signalons qu'alors est valable la proposition suivante:

Si g est une fonction idéale positive, c.-à-d. que l'on a $(q, P) \geqslant 0$ pour tous les polynomes positifs $P(t) \geqslant 0$, alors la fonctionelle qu'elle définit, (g, P) est exprimée par l'intégrale de Stieltjes, c.-à-d. qu'il existe une fonction monotone $\alpha(t)$, telle que l'on a $(g, P) = \int\limits_a^b P(t)\, d\alpha(t)$.

Cette proposition permet parfois de tirer des conclusions quant à la nature de la fonction idéale g.

Remarque 5. Dans la famille B sont comprises pour le cas donné toutes les opérations remplissant les conditions 1)—3).

II. Soit maintenant H_2 un espace de fonctions complexes $x(t)$, à carré sommables avec une période 2π et satisfaisant à la condition $\int\limits_{-\pi}^{+\pi} x(t)\, dt = 0$. Le produit scalaire est $(x, y) = \int\limits_{-\pi}^{+\pi} x(t)\, y(t)\, dt$. Nous pren-

drons ici pous $\mathfrak{B}$ l'ensemble des fonctions $x(t)$ continues et indéfiniment différentiables, faisant partie de H_2.

Introduisons l'opération V comme suit:

$$Vx(t) = -x''(t).$$

Si l'on désigne par $x^{(-1)}(t)$ l'intégrale de la fonction $x(t)$ qui est située dans H_1, c.-à-d. satisfait à la condition $\int_{-\pi}^{\pi} x^{(-1)}(t)\,dt = 0$, l'opération V^{-1} sera:

$$V^{-1}\ x(t) = -x^{(-2)}(t).$$

Prenons pour famille d'opération la famille $B = \{V^\nu\}$; $(\nu = 0, \pm 1, \ldots)$. Il est aisé de vérifier que pour B sont remplies toutes les conditions 1)—7).

Dans $\mathfrak{G}_{V^\nu}$ $(\nu > 0)$ entreront les fonctions $x(t)$ faisant partie de H_2, pour lesquelles l'intégrale $\int_{-\pi}^{+\pi} |x^{(\nu)}(t)|^2\,dt$ est finie. L'ensemble $\mathfrak{G}_{V^{-\nu}}$ peut être réalisé par les fonctions idéales $g(t) = \frac{d^\nu}{dt^\nu}x(t)$, où x est une fonction quelconque prise dans H_2. L'espace $\mathfrak{H}_2$ consistera ici en H_2 et en toutes les fonctions pareilles g. Pour toutes les fonctions provenant de $\mathfrak{H}_2$ sont définies les dérivées et les intégrales de n'importe quel ordre $g^{(\nu)}$ et $g^{(-\nu)}$, qui à leur tour sont des éléments de $\mathfrak{H}_2$. Dans ces conditions, pourvu que ν soit assez grand, $g^{(-\nu)}$ sera certainement situé dans H_2. Si $h \in \mathfrak{G}_{V^\nu}$ et $g \in \mathfrak{G}_{V^{-\nu}}$, le produit scalaire (h, g) est défini. Cela étant, la formule d'intégration par parties est valable: $(h, g) = -(h', g^{(-1)}) = -(h^{(-1)}, g^1)$. Nous le désignerons quelquefois par $(h, g) = \int_{-\pi}^{+\pi} h(t)\,g(t)\,dt$.

Remarque 1. Observons que l'espace prolongé $\mathfrak{H}_2$, ici construit, aurait pu être obtenu aussi bien au paragraphe I. Ainsi, en prenant dans la remarque 3 pour système orthogonal le système de fonctions $\{e^{ikt}\}$ $(k = \pm 1, \pm 2, \ldots)$, et pour γ_k les suites $\left\{\frac{1}{k^\nu}\right\}$ $(\nu = 0, \pm 1, \pm 2, \ldots)$, nous serions arrivés au même résultat. Seulement, l'espace $\mathfrak{H}_2$ se serait trouvé réalisé par des fonctions $g(t)$, données par des séries formelles $g(t) \simeq \sum_{k=\pm 1}^{\pm\infty} a_k e^{ikt}$, où les nombres a_k ne croissent pas plus rapidement qu'un certain k^ν. Les opérations ordinaires effectuées avec ces séries, répondraient aux opérations correspondantes sur les fonctions g.

En particulier, l'opération Cx, envisagée dans I (rem. 2), représente tout simplement le produit des fonctions $c(t)$ et $x(t)$.

Remarque 2. Citons un exemple de fonctions idéales. Considérons la fonction suivante $\sigma(t)$ de H_2:

$$\sigma(t) = \sum_{k=1}^{\infty} \frac{\sin kt}{k} = \sum_{k=1}^{\infty} \frac{1}{2i}\frac{1}{k}\left(e^{ikt} - e^{-ikt}\right) = \begin{cases} \dfrac{-\pi - t}{2} & -\pi \leqslant t < 0 \\ \dfrac{\pi - t}{2} & 0 < t \leqslant \pi \\ 0 & t = 0 \end{cases}.$$

Dans ce cas la fonction $\sigma^{(\nu)}(t)$ sera déjà une fonction idéale faisant partie de $\mathfrak{G}_{V^{-\nu}}$ et il lui correspondra la suite

$$\sigma^{(\nu)}(t) \simeq \sum_{k=1}^{\infty} \frac{1}{2i} \frac{1}{k} [(ik)^{\nu} e^{ikt} - (-ik)^{\nu} e^{-ikt}],$$

et cette série converge $V^{-\nu}$ vers la fonction $\sigma^{(\nu)}(t)$.

III. Considérons encore la réalisation suivante de l'espace de Hilbert H_3. Ses éléments sont donnés par les fonctions $x(t)$, données dans $<-\infty, +\infty>$ et pour lesquelles l'intégrale $\int_{-\infty}^{+\infty} |x(t)|^2 dt$ existe. Le produit scalaire est

$$(x_1, x_2) = \int_{-\infty}^{+\infty} x_1(t) \cdot \overline{x_2(t)}\, dt.$$

Prenons pour $\mathfrak{B}$ un ensemble de fonctions à carré sommables égales à zéro partout en dehors d'un certain intervalle fini.

À chaque fonction $b(t)$, mesurable et bornée dans tout intervalle fini:

$$0 < m_L \leqslant b(t) \leqslant M_L < +\infty, \text{ où } -L \leqslant t \leqslant L,$$

correspond une opération B, définie dans $\mathfrak{B}$.

$$Bx = b(t)\, x(t).$$

Prenons pour B un ensemble de telles opérations $B = \{B\}$.

Alors, ici également, seront remplies les conditions 1)—7). Il est aisé de vérifier qu'ici, en effectuant le prolongement, les éléments idéaux peuvent être réalisés par des fonctions $g(t)$ pour lesquelles l'intégrale $\int_{-L}^{+L} |g(t)|^2 dt$ existe dans tout intervalle fini.

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MATHÉMATIQUES

THÉORIE MATHÉMATIQUE DU COUPLE THERMOÉLECTRIQUE

Par A. TICHONOV (TYCHONOFF)

(*Présenté par S. N. Bernstein, de l'Académie, le 21. VIII. 1935*)

Tout appareil qu'on introduit dans un champ physique pour en mesurer l'intensité provoque une certaine altération de ce champ. Notre appareil nous donne donc l'intensité du champ modifié et non pas celle du champ initial qui nous intéresse en réalité.

Quand on mesure la température à l'aide du couple thermoélectrique une partie de la chaleur passe dans le couple, d'ou provient une certaine altération du champ thermique (l'influence de ce fait est d'autant plus grande que la conductibilité du corps est plus faible). Ce phénomène purement thermique se complique par le fait que le courant électrique produit un dégagement de chaleur en passant par la soudure du couple thermoélectrique (effet de Peltier) de même qu'en passant le long du fil (chaleur de Joule).

Nous voulons établir la relation entre la température $u^*(t)$ observée et la température $\overline{u}^*(t)$ qui existerait en ce point sans l'altération causée par les facteurs susmentionnés: 1) l'état thermique initial du couple et l'écoulement de chaleur qui s'y dirige; 2) l'effet de Peltier; 3) le dégagement de la chaleur de Joule.

Soit à mesurer la température d'un fil homogène infini, le couple thermoélectrique se composant de deux fils infinis homogènes (mais différents) dont les extremités soient soudées; l'une d'entre elles (celle qui se trouve à l'infini) maintenue à une température constante (zéro), et la seconde mise à un moment $t = 0$ en contact avec le point dont on veut savoir la température.

Nous ferons de plus les suppositions suivantes:

1. Si la différence de température des deux soudures est u il se produit dans le couple un courant électrique d'intensité $i = \varphi(u)$.

2. Le courant d'intensité i passant par la soudure durant le temps Δt produit une certaine quantité de chaleur $Q = F(i) \cdot \Delta t$ (effet de Peltier).

3. Le courant d'intensité i passant par le premier fil du couple durant le temps Δt produit une quantité de chaleur égale à $f_1(i)\,\Delta t$, et celui passant par le second fil la quantité $f_2(i) \cdot \Delta t$.

Ordinairement on admet que ces fonctions sont de la forme de

$$\varphi(u) = \alpha \cdot u; \quad F(i) = \beta \cdot i; \quad f_1(i) = \gamma_1 \cdot i^2; \quad f_2(i) = \gamma_2 \cdot i^2;$$

α, β, γ_1, γ_2 étant des constantes. Mais nous n'avons pas besoin de cette restriction et supposons seulement que ce sont des fonctions continues de leurs arguments.

Les équations de la distribution de la chaleur dans les fils du couple thermoélectrique sont

$$k_j \frac{\partial^2 u_j(x, t)}{\partial x^2} + f_j\left[\varphi\left[u_2(0, t)\right]\right] = c_j \rho_j \frac{\partial u_j(x, t)}{\partial t}, \qquad j = 1, 2) \quad (1), (2)$$

k_1, k_2, c_1, c_2, ρ_1, ρ_2 étant des constantes physiques.

Soit $u(x, t)$ la température qu'aurait le fil sans l'altération due à l'introduction du couple. Cette fonction pourrait être définie par les valeurs $u(x, 0)$ qu'elle prend au moment quand on commence la mesure, et par l'équation de la chaleur

$$k \frac{\partial^2 u(x, t)}{\partial x^2} = c\rho \frac{\partial u(x, t)}{\partial t}, \quad (3)$$

où k, c, ρ sont des constantes du fil.

La température réelle du fil $u(x, t)$ satisfait à la même équation (3) pour tous les $x \neq 0$; pour $x = 0$ (au point de la mesure) il se produit un écoulement de chaleur dans le couple. Soit $q_0(t)$ le flux à travers le point $x = 0$; nous avons

$$q_0(t) = \lim_{\varepsilon \to 0} k\left(\frac{\partial u(\varepsilon, t)}{\partial x} - \frac{\partial u(-\varepsilon, t)}{\partial x}\right). \quad (4)$$

Les conditions par lesquelles sont liées les températures du fil et du couple thermoélectrique se composent de deux parties: premièrement il faut égaler les températures du fil et du couple en leur point de contact; en second lieu, il faut évaluer les quantités de chaleur qui affluent en ce point et qui en partent.

Ces conditions nous donnent:

$$u_1(0, t) = u_2(0, t) = u(0, t)\left(= u^*(t)\right); \quad (5)$$

$$q_0(t) + Q(t) = q_1(t) + q_2(t), \quad (6)$$

où

$$q_j(t) = -k_j \frac{\partial u_j(x, t)}{\partial x}\Big|_{x=0} \qquad j = 1, 2) \quad (7), (8)$$

sont les flux de chaleur dans les fils du couple et

$$Q(t) = F\left[\varphi\left[u^*(t)\right]\right] \quad (9)$$

la chaleur dégagée dans la soudure.

De plus on doit satisfaire aux conditions initiales

$$u(x, 0) = \overline{u(x, 0)}; \quad u_1(x, 0) = \psi_1(x); \quad u_2(x, 0) = \psi_2(x), \quad (10)$$

où $\psi_1(x)$, $\psi_2(x)$ sont des fonctions données caractérisant l'état thermique initial du couple thermoélectrique.

Nous avons donc à résoudre le système (A) d'équations (1), (2), (3)

$$\left.\begin{aligned} k \frac{\partial^2 u(x, t)}{\partial x^2} - c\rho \frac{\partial u(x, t)}{\partial t} &= 0; \\ k_1 \frac{\partial^2 u_1(x, t)}{\partial x^2} - c_1\rho_1 \frac{\partial u_1(x, t)}{\partial t} &= -f_1\left[\varphi\left[u_1(0, t)\right]\right]; \\ k_2 \frac{\partial^2 u_2(x, t)}{\partial x^2} - c_2\rho_2 \frac{\partial u_2(x, t)}{\partial t} &= -f_2\left[\varphi\left[u_2(0, t)\right]\right] \end{aligned}\right\} \quad (A)$$

avec les conditions

$$\left.\begin{aligned} &u(0, t) = u_1(0, t) = u_2(0, t); \\ &q_0(t) + Q(t) = q_1(t) + q_2(t), \end{aligned}\right\} \qquad \text{(B)}$$

les fonctions $q_0(t)$, $q_1(t)$, $q_2(t)$, $Q(t)$ étant définies par les formules (4), (7), (8), (9) et les conditions initiales (10)

$$u(x, 0) = \overline{u(x, 0)}; \quad u_1(x, 0) = \psi_1(x); \quad u_2(x, 0) = \psi_2(x). \qquad \text{(C)}$$

Si l'on prend

$$u(x, t) = \overline{u(x, t)} = \frac{1}{2\sqrt{\pi}} \int_0^t \frac{a}{\sqrt{t-\tau}} e^{-\frac{x^2}{4a^2(t-\tau)}} \frac{q_0(t)}{k} d\tau, \quad \left(a^2 = \frac{k}{c\rho}\right), \qquad (11)$$

la fonction $u(x, t)$ satisfait à l'équation (3), à la condition (4) et possède les valeurs initiales requises.

Les fonctions $u_1(x, t)$ et $u_2(x, t)$ sont à prendre sous la forme

$$u_j = \frac{1}{2\sqrt{\pi}} \int_0^t \int_0^\infty \frac{a_j}{\sqrt{t-\tau}} \left\{ e^{-\frac{(x+\xi)^2}{4a_j^2(t-\tau)}} - e^{-\frac{(x+\xi)^2}{4a_j^2(t-\tau)}} \right\} \frac{1}{c_j\rho_j} f_j\left[\varphi[u_j(0, \tau)]\right] d\xi d\tau +$$

$$+ \frac{1}{2\sqrt{\pi}} \int_0^t \frac{a_j}{\sqrt{t-\tau}} e^{-\frac{x^2}{4a_j^2(t-\tau)}} \mu_j(\tau)\, d\tau +$$

$$+ \frac{1}{2\sqrt{\pi}} \int_0^\infty \frac{a_j}{\sqrt{t}} \left\{ e^{-\frac{(x-\xi)^2}{4a_j^2 \cdot t}} - e^{-\frac{(x+\xi)^2}{4a_j^2 \cdot t}} \right\} \psi_j(\xi)\, d\xi. \quad \left(a_j^2 = \frac{k_j}{c_j\rho_j}\right) \qquad (12)$$

où $\mu_j(t)$ $(j = 1, 2)$ sont des fonctions inconnues; pour que la condition (5) soit remplie, il faut que l'on ait

$$a_1 \cdot \frac{1}{2\sqrt{\pi}} \int_0^t \frac{\mu_1(\tau)}{\sqrt{t-\tau}} d\tau = a_2 \cdot \frac{1}{2\sqrt{\pi}} \int_0^t \frac{\mu_2(\tau)}{\sqrt{t-\tau}} d\tau =$$

$$= \overline{u(0, t)} - \frac{a}{k} \cdot \frac{1}{2\sqrt{\pi}} \int_0^t \frac{q_0(t)}{\sqrt{t-\tau}} d\tau \left(= u^*(t)\right). \qquad (13)$$

Évaluons $q_l(t)$ $(l = 1, 2)$

$$q_j(t) = -k_j \left.\frac{\partial u_j(x, t)}{\partial x}\right|_{x=0} =$$

$$= -\frac{k_j}{2\sqrt{\pi}} \int_0^t \int_0^\infty \frac{\xi}{2a_j(t-\tau)^{3/2}} e^{-\frac{\xi^2}{4a_j^2(t-\tau)}} \frac{1}{c_j\rho_j} f_j\left[\varphi[u_j(0, \tau)]\right] d\xi d\tau +$$

$$+ \frac{1}{2} k_j \mu_j - \frac{k_j}{2\sqrt{\pi}} \int_0^\infty \frac{\xi}{2a_j t^{3/2}} e^{-\frac{\xi^2}{4a_j^2 t}} \psi_j(\xi)\, d\xi = -\frac{k_j a_j}{c_j\rho_j} \cdot \frac{1}{2\sqrt{\pi}} \int_0^t \frac{f_j\left[\varphi[u_j(0, \tau)]\right]}{\sqrt{(t-\tau)}} d\tau +$$

$$+ \frac{1}{2} k_j \mu_j(t) - \frac{k_j}{2\sqrt{\pi}} \int_0^\infty \frac{\xi}{2a_j t^{3/2}} e^{-\frac{\xi^2}{4a_j^2 \tau}} \psi(\xi)\, d\xi. \qquad (14)$$

Donc l'équation (5) peut s'écrire comme suit:

$$q_0(t) = \frac{1}{2} k_1\mu_1(t) + \frac{1}{2} k_2\mu_2(t) - \frac{k_1a_1}{c_1\rho_1} \cdot \frac{1}{2\sqrt{\pi}} \int_0^t \frac{f_1\left[\varphi\left[u(0,t)\right]\right]}{\sqrt{t-\tau}} d\tau -$$

$$- \frac{k_2a_2}{c_2\rho_2} \cdot \frac{1}{2\sqrt{\pi}} \int_0^t \frac{f_2\left[\varphi\left[u(0,t)\right]\right]}{\sqrt{t-\tau}} d\tau - \frac{k_1}{2\sqrt{\pi}} \int_0^\infty \frac{\xi}{2a_1t} e^{-\frac{\xi^2}{4a_1^2t}} \psi_1(\xi)\, d\xi -$$

$$- \frac{k_2}{2\sqrt{\pi}} \int_0^\infty \frac{\xi}{2a_2t} e^{-\frac{\xi^2}{4a_2^2t}} \psi_2(\xi)\, d\xi - F\left[\varphi\left[u(0,t)\right]\right]. \qquad (15)$$

Nous allons maintenant éliminer des équations (13) et (15) les fonctions inconnues $\mu_1(t)$, $\mu_2(t)$, $q_1(t)$, $q_2(t)$.

Nous utiliserons à cet effet les deux remarques suivantes:

(a)
$$\int_0^T \frac{1}{\sqrt{T-t}} \left\{ \int_0^t \frac{f\left[\varphi\left[u(0,\tau)\right]\right]}{\sqrt{t-\tau}} d\tau \right\} dt =$$

$$= \int_0^T f\left[\varphi\left[u(0,\tau)\right]\right] \left\{ \int_\tau^t \frac{dt}{\sqrt{(T-t)(t-\tau)}} \right\} d\tau = \pi \int_0^T f\left[\varphi\left[u(0,\tau)\right]\right] d\tau.$$

(b) Pour toute fonction bornée satisfaisant à l'équation de la conductibilité calorifique

$$a^2 \frac{\partial^2 v(x,t)}{\partial x^2} = \frac{\partial v(x,t)}{\partial t}$$

on a

$$v(x,t) = \frac{1}{2\sqrt{\pi}} \int_0^t \frac{a}{\sqrt{t-\tau}} e^{-\frac{x^2}{4a^2(t-\tau)}} \left[-2 \frac{\partial v(0,\tau)}{\partial x} \right] d\tau.$$

Cela posé, considérons

$$v(x,t) = \frac{a}{\sqrt{t}} e^{-\frac{(x+\xi)^2}{4a^2t}}; \qquad (\xi > 0)$$

cette fonction est bornée et satisfait à l'équation de la conductibilité calorifique, donc

$$\frac{a}{\sqrt{t}} e^{-\frac{(x+\xi)^2}{4a^2t}} = \frac{1}{2\sqrt{\pi}} \int_0^t \frac{a}{\sqrt{t-\tau}} e^{-\frac{x^2}{4a^2(t-\tau)}} \left[\frac{\xi}{a\tau^{3/2}} e^{-\frac{\xi^2}{4a^2\tau}} \right] d\tau.$$

En particulier pour $x = 0$ nous obtenons

$$\frac{a}{\sqrt{t}} e^{-\frac{\xi^2}{4a^2t}} = \frac{1}{2\sqrt{\pi}} \int_0^t \frac{a}{\sqrt{t-\tau}} \left[\frac{\xi}{a\tau^{3/2}} e^{-\frac{\xi^2}{4a^2\tau}} \right] d\tau.$$

De là on conclut que

$$\int_0^T \frac{1}{\sqrt{T-t}} \left\{ \frac{1}{2\sqrt{\pi}} \int_0^\infty \frac{\xi}{2at^{3/2}} e^{-\frac{\xi^2}{4a^2t}} \psi(\xi)\, d\xi \right\} d\tau = \frac{1}{2} \int_0^\infty \frac{1}{\sqrt{T}} e^{-\frac{\xi^2}{4a^2T}} \psi(\xi)\, d\xi.$$

En utilisant le dernier membre de l'égalité (13) et les remarques (a), (b), on voit immédiatement

$$\overline{u(0,t)} = \overline{u^*}(t) = u(0,t) + \frac{a}{k}\cdot\frac{1}{2\sqrt{\pi}}\int_0^t \frac{q_0(\tau)}{\sqrt{t-\tau}}\,d\tau =$$

$$= u(0,t) + \frac{ak_1}{2k}\cdot\frac{1}{2\sqrt{\pi}}\int_0^t \frac{\mu_1(\tau)}{\sqrt{t-\tau}}\,d\tau + \frac{ak_2}{2k}\frac{1}{2\sqrt{\pi}}\int_0^t \frac{\mu_2(\tau)}{\sqrt{t-\tau}}\,d\tau -$$

$$-\frac{aa_1^2}{k}\cdot\frac{1}{2\sqrt{\pi}}\int_0^t f_1\Big[\varphi[u(0,\tau)]\Big]\,d\tau - \frac{aa_2^2}{k}\cdot\frac{1}{2\sqrt{\pi}}\int_0^t f_2\Big[\varphi[u(0,\tau)]\Big]\,d\tau -$$

$$-\frac{ak_1}{k}\cdot\frac{1}{2\sqrt{\pi}}\int_0^\infty \frac{1}{\sqrt{t}}e^{-\frac{\xi^2}{2a_1^2t}}\phi_1(\xi)\,d\xi - \frac{ak_2}{k}\cdot\frac{1}{2\sqrt{\pi}}\int_0^\infty \frac{1}{\sqrt{t}}e^{-\frac{\xi^2}{4a_2^2t}}\phi_2(\xi)\,d\xi -$$

$$-\frac{a}{k}\cdot\frac{1}{2\sqrt{\pi}}\int_0^t \frac{F\Big[\varphi[u(0,\tau)]\Big]}{\sqrt{t-\tau}}\,d\tau$$

ou, en définitive,

$$\overline{u^*}(t) = u^*(t)\Big[1 + \frac{a}{2k}\Big(\frac{k_1}{a_1} + \frac{k_2}{a_2}\Big)\Big] - \frac{aa_1^2}{k}\cdot\frac{1}{2\sqrt{\pi}}\int_0^t f_1\Big[\varphi[u^*(\tau)]\Big]\,d\tau -$$

$$-\frac{aa_2^2}{k}\cdot\frac{1}{2\sqrt{\pi}}\int_0^t f_2\Big[\varphi[u^*(\tau)]\Big]\,d\tau - \frac{ak_1}{k}\cdot\frac{1}{2\sqrt{\pi}}\int_0^\infty \frac{1}{\sqrt{t}}e^{-\frac{\xi^2}{4a_1^2t}}\phi_1(\xi)\,d\xi -$$

$$-\frac{ak_2}{k}\cdot\frac{1}{2\sqrt{\pi}}\int_0^\infty \frac{1}{\sqrt{t}}e^{-\frac{\xi^2}{4a_2^2t}}\phi_2(\xi)\,d\xi - \frac{a}{k}\cdot\frac{1}{2\sqrt{\pi}}\int_0^t \frac{F\Big[\varphi[u^*(\tau)]\Big]}{\sqrt{t-\tau}}\,d\tau.$$

Nous avons déjà mentionné que les fonctions φ, f_1, f_2, F ont la forme de:

$$\varphi(u) = \alpha\cdot u; \quad F(i) = \beta\cdot i; \quad f_1(i) = \gamma_1\cdot i^2; \quad f_2(i) = \gamma_2\cdot i^2.$$

Dans ce cas on obtient

$$\overline{u^*}(t) = u^*(t)\Big[1 + \frac{a}{2k}\Big(\frac{k_1}{a_1} + \frac{k_2}{a_2}\Big)\Big] + \frac{a\alpha^2}{2\sqrt{\pi}\cdot k}(\gamma_1 a_1^2 + \gamma_2 a_2^2)\int_0^t u^*(\tau)^2\,d\tau +$$

$$+\frac{a}{k}\cdot\frac{\alpha\cdot\beta}{2\sqrt{\pi}}\int_0^t \frac{u^*(\tau)}{\sqrt{t-\tau}}\,d\tau + \frac{a}{k}\left\{\frac{k_1}{a_1}\cdot\frac{1}{2\sqrt{\pi}}\int_0^\infty \frac{a_1}{\sqrt{t}}e^{-\frac{\xi^2}{4a_1^2t}}\phi_1(\xi)\,d\xi -\right.$$

$$\left. -\frac{k_2}{a_2}\cdot\frac{1}{2\sqrt{\pi}}\int_0^\infty \frac{a_2}{\sqrt{t}}e^{-\frac{\xi^2}{4a_2^2t}}\phi_2(\xi)\,d\xi\right\}.$$

C'est bien la forme de la fonction $\overline{u^*}(t)$ qui représente la température cherchée en termes de $u^*(t)$, température mesurée. Le premier membre dépend seulement de la perte de chaleur qui aurait lieu si la tempéra-

ture initiale du couple thermoélectrique était égale à zéro, c.-à-d. à la température de la soudure maintenue à un niveau fixe; le second dépend de la chaleur de Joule, le troisième de l'effet de Peltier, et enfin le quatrième de la distribution initiale de la chaleur dans le couple.

En particulier, si le fil dont on mesure la température a les mêmes qualités thermiques que le couple, c.-à-d. si leurs constantes a, k sont égales et si les distributions initiales sont

$$\phi_1(\xi) = \phi_2(\xi) = 0,$$

c.-à-d. si la température dans tous les points du couple thermoélectrique est égale à celle de la soudure à l'infini, et si l'on néglige les effets de Peltier et de Joule,—alors on a

$$\overline{u}^*(t) = u^*(t) \cdot 2.$$

Done, dans ces conditions s'établit instantanément une température constante égale à 50% de celle qu'on veut déterminer.

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PHYSIK

ÜBER DEN EINFLUSS DES DRUCKES AUF DIE ENTLADUNGS-STRAHLUNG IN QUECKSILBERDÄMPFEN

Von V. FABRIKANT, F. BUTAJEVA und J. CIRG

(Vorgelegt von S. I. Wawilow, Mitglied der Akademie, d. 4. VIII. 1935)

In der vorhergehenden Mitteilung sprachen wir die Vermutung aus, dass das Fehlen eines Minimums für die Strahlungsintensität bei der Entladung in Quecksilberdämpfen bei ansteigendem Druck auf den Einfluss eines indifferenten Gases zurückgeführt werden kann (1). Hierbei gingen wir von den Ergebnissen aus, die wir für Kadmiumentladungen erhielten. Die an Quecksilberentladungen angestellten Messungen bestätigten obige Vermutung.

Die von uns in Gegenwart von Argon angestellten Intensitätsmessungen der sichtbaren Entladungsstrahlung in Quecksilberdämpfen in ihrer Abhängigkeit vom Druck ergaben das gleiche Resultat wie die Krefftschen (2): mit anderen Worten, die Intensität verbleibt bis zu einem gewissen Drucke konstant, um sodann schnell zu sinken.

Nun ergibt sich aber bei Entladungen in Quecksilberdämpfen in Gegenwart geringer Neonmengen (2 mm Druck) ein in Abhängigkeit vom Druck scharf ausgeprägtes Intensitätsminimum. Je niedriger der Druck des Neons, umso schärfer das Minimum.

Somit gleicht sich bei der Entladung in Quecksilber- wie auch in Kadmiumdämpfen die Abhängigkeit der Strahlungsintensität vom Druck aus, wobei Argon einen stärkeren Einlfuss ausübt als Neon. In der vorhergehenden Mitteilung kamen wir auf Grund des ermittelten Unterschiedes im Lauf der einzelnen sichtbaren Tripletlinien zum Schluss, dass die Reabsorption des sichtbaren Triplets bei der Kadmiumentladungsstrahlung unter niedrigem Druck einen hohen Wert erreichen muss (1). Offenbar müsste man einen ähnlichen Effekt auch bei der Quecksilberentladung wahrnehmen.

Um diese Behauptung auf direktem Wege nachprüfen zu können, stellten wir Absorptionsmessungen an Quecksilberentladungen unter niedrigem Druck (10^{-2} mm Hg) an. Damit die Strahlungsquelle Linien von der ähnlichen Form wie die absorbierende Entladungssäule liefere, bedienten wir uns der Spiegelmethode*. Als Strahlungsquelle benutzten wir die imaginäre Abbildung derjenigen Entladungsröhre, welche zur Messung der

* Diese Methode versuchten wir zuerst, in Gemeinschaft mit V. L. Pulver, im J. 1933 zur Untersuchung von Entladungen in Neon anzuwenden.

Absorption diente. Diese «Autokollimationsmethode» gestattet nur die Absorptionswerte abzuschätzen, sowie auch den Verlauf der Absorption in Abhängigkeit von verschiedenen Bedingungen zu bestimmen.

Unsere Messungen ergaben, dass die Absorption der Linien 5464 Å und 4358 Å grösser ist als die der Linien 4960 Å und 5770/90 Å, und dass die Absorption der Linie 4358 Å mehr als 65% beträgt! Hierbei ist die Absorption der Linie 4358 Å grösser als die der Linie 5464 Å. Diese Tatsache ist insofern von Interesse, als die Linie 5464 Å am metastabilen Niveau grenzt, was bei der Linie 4358 Å nicht der Fall ist. Wir sehen, dass die Absorption bei Niederdruckentladung ihrer Grösse nach solche bei Hochdruckentladung übertreffen kann [3].

Es ist einleuchtend, dass die erheblichen Reabsorptionswerte diejenigen Abweichungen im Intensitätsverhältnis des sichtbaren Quecksilbertriplets von dem auf Grund der Intensitätsregeln festgestellten Verhältnis erklären, welche bei einer Anzahl von Niederdruckentladungen beobachtet wurden [4] (ausführlicher darüber s. unsere Abhandlung im Bulletin des Elektrotechnischen Instituts, 1935).

Elektrotechnisches Institut.
Moskau.

Eingegangen
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CHEMISTRY

THE EFFECT OF THE WATER VAPOUR PRESSURE ON THE RATE OF THE INTERACTION BETWEEN IODINE AND METALS (IRON AND COPPER)

By A. V. SOLOVJEV

(Communicated by V. A. Kistiakovskij. Member of the Academy. 23. VIII. 1935)

In investigating the reactions between solid bodies and gases it is usually assumed that, if a compact layer of solid compounds be formed during the course of the reaction, further action depends upon the diffusion of the gases through this layer. This process has found mathematical expression from the standpoint of molecular kinetics in the parabolic equation ([1], [2], [3], [4], [5]) $y^2 2kt$, where y is either the thickness of the film, or the increase in weight, or the quantity of reacting substances, and t is the time.

In investigating the oxidation of metals, it is possible to limit oneself, as Pilling, Bedworth and others did, to a consideration of the surface layer produced during the reaction, but in investigating the action of gases on metals that have been exposed to the air, it is necessary to allow for the already existing film of oxides on the surface of the metal, as has been shown by Academician V. A. Kistiakovskij ([6]), who has proposed a new method of investigating the «solidation» of a film of iron under the action of iodine.

Consequently, in the case described below, this process must consist in the diffusion of the gas through the primary film of oxides, then the destruction of this film, and, lastly, the formation of a layer of new compounds. If a liquid absorbed from the gaseous phase condenses on the surface of the metal, there occurs both chemical and electrochemical destruction of the metal by the local currents arising between the metal and its film or between its inclusions.

Academician V. A. Kistiakovskij proposed a new method of investigating the phenomena of the corrosion of iron by accelerating the process with iodine vapour. We subsequently applied this method to other metals.

For the purpose of studying the dynamics of the complicated processes of the interaction between metals and iodine, we elaborated a gravimetrical method in which the changes in weight were recorded by means of a special apparatus designed after the type of the Nernst balance (changes in weight could be noted by changes in the degree of curvature of a fine glass thread; quartz torsion balance may also be used). This apparatus was placed under a bell jar enclosing a definite gaseous phase. Where the use of this apparatus was precluded by rapid condensation of moisture, the dynamics of the process were characterized by changes in the weight of strips of iden-

tical chemical composition and dimensions, which were exposed for a definite time to the gaseous phase and then freed from the chemical products of this interaction. Control of the vapour pressure was effected by means of phosphoric acid of corresponding concentrations.

Kohlschütter and Krähenbuhl (7) found that, on acting with halogens on copper and silver, deviations occur from the parabolic equation instead of accordance that could have been expected on the basis of Tammann's experiments (1). Our data confirmed the data both of Kohlschütter and Krähenbuhl and of Tammann, for the parabolic equation does not always hold exactly, but depends upon the water vapour pressure, although the action of iodine on copper is always expressed by curves directed to the axis of time.

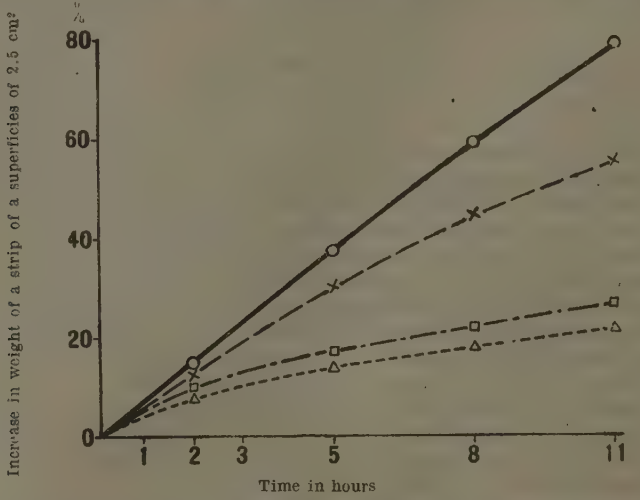

Fig. 1. For the curve consisting of small circles, P (water vapour pressure)=12.699 mm; for that consisting of crosses, P=12.07 mm; for that consisting of triangles, P=11.18 mm; for that consisting of squares, P=6.18 mm. The gaseous phase is saturated with iodine vapours. $t°$=15° C. Kahlbaum electrolytic copper.

The optimum conditions under which the course of the reaction with copper follows the parabolic equation is a water vapour pressure of 6.18 mm at a temperature of 15° C. At pressures above 11.18 mm, the interaction of the reacting substances is more intense as a result of increased diffusion through the less compact layer of newly formed compounds, as is shown by the following graph (see Fig. 1).

Here the axis of abscisses expresses the time and that of ordinates the increase in weight (the percentages of the increase in weight of copper strips of a superficies of 2.5 cm² are measured along the y axis), P is the pressure of water vapour in mm of mercury.

This phenomenon is explained by the «solidation» (coagulation, formation of crystals, crystallization, conversion from one phase into another) of the primary film in the beginning of the process, which leads to the reaction of the iodine and the active (film-free) sections of metal followed by the formation of a layer of compounds. The subsequent process depends upon the physical qualities of these compounds. Since on the transition from a moist gaseous phase to dry one there occurs a solidification of the film,

a contraction of the pores, the process is dynamic and may be expressed by the equation of Evans (11), more general, than the one mentioned above, viz.

$$\frac{dy}{dt} = \frac{c_0 k_0 k_1}{k_0 + y k_1},$$

which includes, as a special case, the parabola (if k_0 is very small as compared with yk, $\frac{dy}{dt} = \frac{c_0 k_0}{y}$ or $y^2 = 2 c_0 k_0 t + \text{const}$), and the straight line (if yk is very small as compared with k_0, $\frac{dy}{dt} = c_0 k$)

Here $\frac{dy}{dt}$ expresses the increase in weight, k_0 is the constant of the film's permeability, k_1 is the chemical constant, and c_0 is the gas concentration between the internal surface of the layer formed on the metal during the

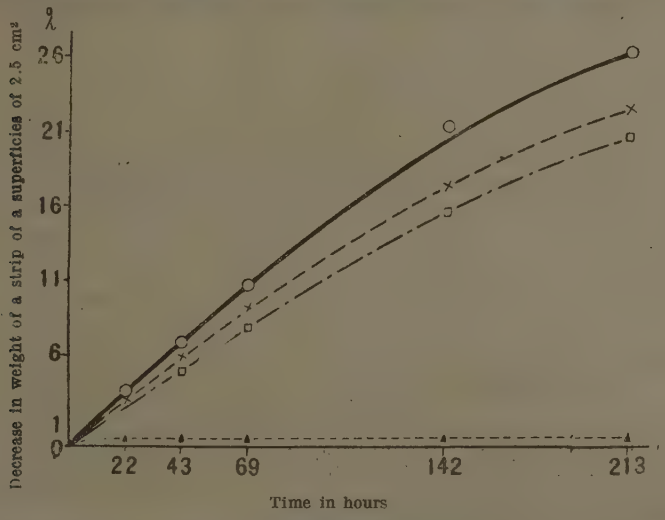

Fig. 2. Same symbols as in Fig. 1. Chemical composition of iron: 0.05 C, traces of Si, traces of P, 0.032 Mn, 0.04 S.

reaction and the metal itself. In a moist atmosphere, separation of the film from the surface of the metal is periodically observed to occur. This leads to an increased action of the iodine, which is reflected in the graph by certain fluctuations.

The interaction of iodine and iron is more complicated than that of iodine and copper, for in the case of iron a condensed liquid forms on its surface due to the absorption of moisture from the gaseous phase by the highly hygroscopic ferrous iodide formed during the reaction. This sets up favourable conditions for the appearance of local electric currents. Nevertheless, the general course of the process is also expressed by curves directed to the axis of time (see Fig. 2), although at the beginning of the reaction the rate of the interaction is expressed by a straight line.

If the water vapour pressure be decreased (as against that of saturation) by 6.5 mm (over 84.63% H_3PO_4), the reaction slackens considerably.

The initial process of the action of iodine on iron is considerably affected by the different rates of solidation of the film dependent on the water vapour

pressure. The highest rate of solidation of the film corresponds for iron to a decrease in water vapour pressure of 0.5—0.6 mm of mercury. This fact explains the phenomenon observed by us ([8],[9]) for iron and still earlier by Vernon ([10]) for copper, viz. that the highest rate of corrosion under atmospheric conditions does not correspond to complete saturation of the atmosphere with water vapour, as could have been expected, but to a somewhat lower value. Nevertheless for long periods of time it holds that with strong reagents (iodine in this case) the quantity of substances involved in the reaction

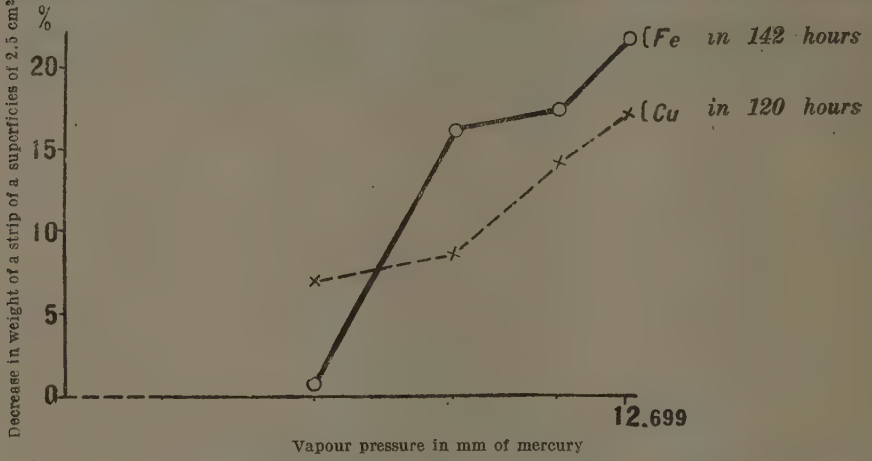

Fig. 3. For the first point on the X axis, P=6.18 mm; for the second, P=11.18 mm; for the third, P=12.07 mm; for the fourth, P=12.699 mm.

is the greater the higher the humidity of the atmosphere, since the period of solidation of the film is not long and the process depends on the quantity of moisture condensed and, for Cu, on the porosity of the layer formed, as is shown in the following graph (see Fig. 3).

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CHEMISTRY

THE INTERACTION BETWEEN ALUMINIUM AND IODINE IN AN AIR ATMOSPHERE OF DIFFERENT HUMIDITY

By A. V. SOLOVJEV

(*Communicated by V. A. Kistiakovskij, Member of the Academy. 23. VIII. 1935*)

There are two maxima for the interaction of iodine and aluminium in the presence of air of different humidity: one of them corresponds to full saturation of the atmosphere with water vapour, that is, according to Renier, to a pressure of 12.699 mm of mercury at 15° C, and the other to the water

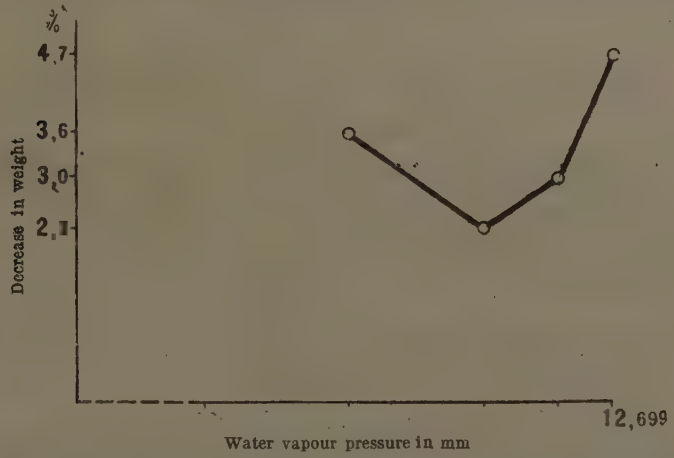

Fig. 1. For the first point on the X axis, P=6.18 mm; for the second point, P=11.18 mm; for the third point, P=12.07 mm; for the fourth point, P=12.699 mm.

vapour pressure over 84.63% H_3PO_4, that is, according to our computation made after the formula $\frac{\Delta p}{p}=\frac{in}{N+in}$ to 6.18 mm of mercury. In a dry atmosphere (over dry $CaCl_2$) the action again decreases (this part of the curve is not shown in the graph, Fig. 1).

The process of the solidation of the film may be easily studied in the case of aluminium, as this process continues for several days when iodine acts upon aluminium at a pressure of water vapour equal to 6.18 mm of mercury and can thus be observed with the naked eye.

On decreasing the pressure of water vapour by 6.5 mm, the initial process is determined exclusively by the rate of the solidation of the aluminium film and that of the reaction of iodine and the film-free sections of the metal, while on decreasing the pressure of water vapour by as much as 1.5 mm, there occurs a condensation of the liqiud in addition to the solidation of the film, and consequently both chemical and electrochemical destruction of the metal ensues.

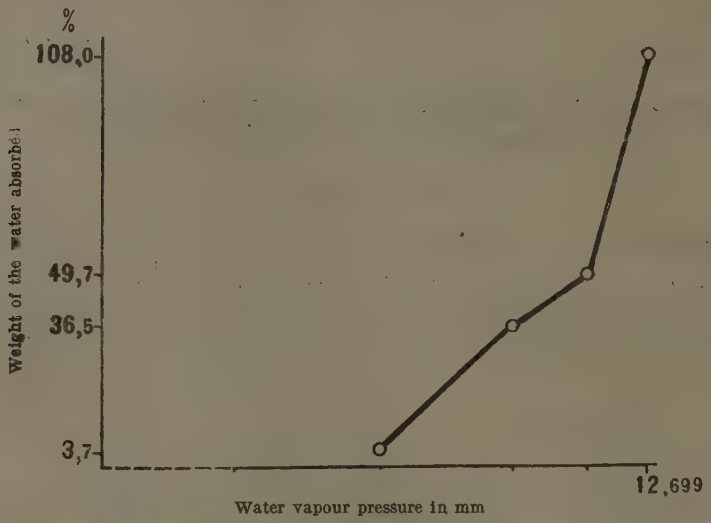

Fig. 2. For the first point on X axis P=6.18 mm; for the second point, P=11.18 mm; for the third point, P=12.07 mm; for the fourth point, P=12.699 mm.

The graph shows that moisture, on the one hand, retards the process of the solidation of the film and that, on the other hand, the more moisture there is, the quicker will the film, and consequently the metal itself, be dissolved.

The following graph (see Fig. 2) shows the quantity of moisture condensed on the surface of the aluminium strips (of a superficies of 2.5 cm²) as the result of the hygroscopicity of the compounds produced by the reaction in 96 hours, in dependence upon the pressure of water vapour.

The quantity of moisture participating in the reaction at a vapour pressure corresponding to 10.9 mm of mercury is very small. Analysis has shown that under such conditions the main reaction product is AlJ_3.

It is characteristic of the rates of the reaction of aluminium and iodine in the presence of moist air that they are expressed by certain curves directed to the axis of decrease in weight and not to the axis of time, as will be seen in the following graph (see Fig. 3).

Such a course of the corrosion process at a water vapour pressure of over 6.18 mm is due to the increase with time of the amount of the condensed moisture retained on the surface and causing chemical and electrochemical dissolution. The electrolyte is in this case the aluminium iodide solution formed on the surface of the metal in the process of the interaction of Al and J_2.

In the absence of moisture condensed into drops on the surface of aluminium, the process depends on the rate of the liberation of the aluminium

from its film, i. e. on the rate of its solidation. But as the solidation proceed from the centres formed towards the periphery, it is expressed by the area of the circle. This determines the character of the curves.

If there were only one centre of solidation, it might be expected that the process would follow the law of parabola. There are several such centres, how-

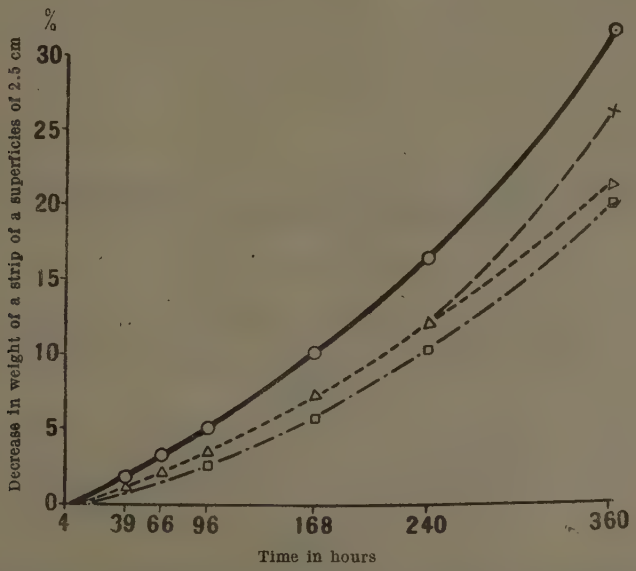

Fig. 3. For the curve consisting of circles, P (water vapour pressure)=12.699 mm; for that consisting of crosses, P=12.07 mm; for that consisting of triangles, P=11.18; for that consisting of squares, P=6.18 mm. The gaseous phase is saturated with iodine vapours, $t°$=15°C. Kahlbaum aluminium.

ever, and on increasing the surface they coalesce; therefore, a deviation from the law of parabola is observed to occur.

In conclusion I wish to express my indebtedness to Academician V. A. Kistiakovskij and senior specialist I. V. Krotov for their advice and valuable suggestions in the course of my work.

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CHEMISTRY

THE SOLUBILITY AND ACTIVITY OF THE HALOGENATES OF SOME BIVALENT METALS

A. POLESSITSKIJ

(Communicated by J. V. Grebenščikov, Member of the Academy, 10. XI. 1935)

I. The Solubility and Activity of $Ba(JO_3)_2$ in Water and in Solutions of Electrolytes

The solubility of $Ba(JO_3)_2$ in water and in solutions of electrolytes at 25° was carefully determined by Harkins and Winninghoff (1) up to concentration of electrolyte as great as 0.2 mol/l. It was interesting to prolong their investigation to much greater concentrations. The solubility of $Ba(JO_3)_2$ at temperatures from 0 to 100° was determined by Trautz and Anschütz (2), but as their result at 25° was much lower than that of Harkins and Winninghoff, it was desired to verify the solubility at all temperatures.

M e t h o d. Recrystallised $BaCl_2.2H_2O$ was precipitated by an equivalent amount of KJO_3; the precipitate was filtered off and washed with hot water, or washed by decantation with large amount of cold water, under strong stirring.

For solubility determinations this salt was stirred with water or electrolytic solution, in a thermostate during 24 hours. After settling the crystals during 1 hour samples were taken with a pipette with a cotton-wool filter. To avoid the adsorption of the salt on the pipette and cotton-wool, the first 3—4 portions of the solution were thrown away.

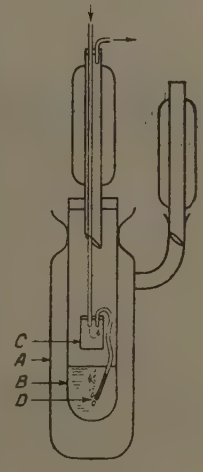

Fig. 1.

The solubility at high temperatures was determined in an apparatus shown in Fig. 1. In the outer tube *A* a liquid was boiling, whose vapour gave a constant temperature (H_2O—100°, C_2H_5OH—78°). The salt was mixed with water in the inner tube *B* by an air current, whose direction is given in the figure with arrows. The samples were taken in a glass *C*, through a tube with a cotton-wool, filter *D* by an air current of the opposite direction.

The samples were analysed by different methods. The solutions in water were analysed with the help of: 1) the evaporation of 200 cm³ of the solution

and the drying at 90°. 2) the determination of Ba as $BaSO_4$ from 200 cm³ of the solution and 3) the iodometric titration of JO_3'. In the last method 1 cm³ of the solution was treated with 25 cm³ H_2O, 5 cm³ 25% HCl, 1 cm³ 10% KJ and titrated with $n/1000$ $Na_2S_2O_3$, with starch as indicator. All these methods gave the same results within ± 3%. The solubility of $Ba(JO_3)_2$ in presence of other electrolytes was determined only by an iodometric titration (method 3).

Results. The solubility of $Ba(JO_3)_2$ in water at different temperatures is given in Table 1 and also in Fig. 2. For comparison, in the same figure a very precise result by Harkins and Winninghoff at 25°, which is very near

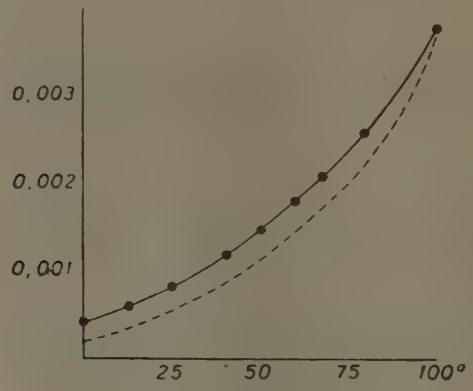

Fig. 2.

to my data, is given, and also the results by Trautz and Anschütz, which are lower than mine at all temperatures. Perhaps their low results depend upon the adsorption of the salt on the pipette and the cotton-wool filter.

Table 1

Solubility of $Ba(JO_3)_2$ in water

$t°$	mg/l	mol/l
0	182	0.000374
15	305	0.000626
25	406.3	0.000833
40	580.0	0.00119
50	727	0.00149
60	873	0.00179
70	1 070	0.00220
78	1 300	0.00267
100	1 840	0.00378

The solubility of $Ba(JO_3)_2$ in concentrated solutions of KNO_3 and $Ca(NO_3)_2$ at 25° is given in table 2 and Fig. 3.

Table 2

Solubility of $Ba(JO_3)_2$ in concentrated solutions of electrolytes at 25°

Electrolyte	Concentration mol/l	Solubility of $Ba(JO_3)_2$		Total ionic strength	Activity coefficient
		mg/l	mol/l		
—	—	406.3	0.000833	0.005	0.988
KNO_3	0.2	833	0.00171	0.410	0.4811
»	1.4	2 050	0.00421	2.825	0.195
»	2.25	2 770	0.00568	4.534	0.145
$Ca(NO_3)_2$	1.36	4 266	0.00874	7.853	0.094
»	2.27	5 196	0.01064	13.68	0.077
»	3.07	6 457	0.01324	18.50	0.0621
»	3.60	6 500	0.01333	21.68	0.0615
»	4.22	5 519	0.01133	25.38	0.0725
»	5.18	4 022	0.00825	31.13	0.0997

In Fig. 3 the solubility of $Ba(JO_3)_2$ is given against total ionic strength of solutions. The extrapolation of this curve to zero ionic strength gives the

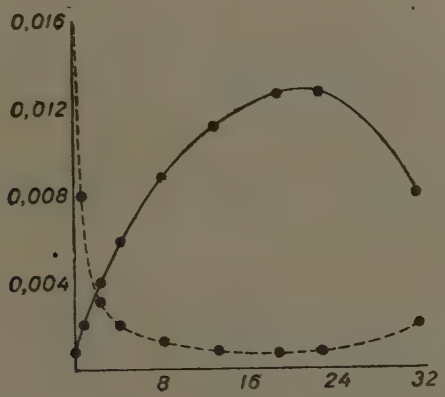

Fig. 3.

«limiting solubility» not distorted by an ionic field. For 25° it is 400.7 mg/l.

The activity coefficients were calculated from the comparison of the limiting solubility with the solubility in the corresponding solutions. The dotted line in fig. 3 gives the activity coefficients against the ionic strength.

Summary

1. The solubility of $Ba(JO_3)$ in water at temperatures from 0° up to 100° was determined.

2. The solubility of $Ba(JO_3)_2$ in concentrated solution of KNO_3 and $Ca(NO_3)_2$ at different concentrations of the electrolytes was determined.

3. The limiting solubility and the activity coefficients at different values of the ionic strength were calculated.

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[2] Trautz u. Anschütz, Z. Phys. Chem. **56**, 238, 1906.

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CHEMISTRY

THE SOLUBILITY AND ACTIVITY OF THE HALOGENATES OF SOME BIVALENT METALS

A. POLESSITSKIJ

(Communicated by J. V. Grebenščikov, Member of the Academy, 10. XI. 1935)

II. The Solubility and Activity of $Pb(JO_3)_2$ in Water and in Solutions of Electrolytes

The solubility of $Pb(JO_3)_2$ in water and in solutions of electrolytes was determined at 25° by Harkins and Winninghoff (1) up to concentrations as great as 0.2 m, and at room temperature (17—20°) by Geilmann and Haltje (2) up to 0.001 m. There are no data for greater concentrations. The solubility in water was determined also at 9.2, 18 and 25.8°. It was of interest to study the solubility of $Pb(JO_3)_2$ in water at different temperatures and in solutions of electrolytes at great concentrations.

M e t h o d. Recrystallised $Pb(NO_3)_2$ was precipitated by an equivalent amount of KJO_3. The precipitate was filtered off and washed with hot water, or washed by decantation with large amount of cold water under strong stirring.

The solubility of $Pb(JO_3)_2$ in water or in electrolytic sloution was determined by the same method as the solubility of $Ba(JO_3)_2$ (4). The analysis by the help of the evaporation of the solution was made only in one case (the solution in water at 25°). In all other experiments was applied the iodometric titration of JO_3^-. For this purpose 10—30 cm³ of the solution were treated with 25 cm H_2O, 5 cm³ 25 % HCl, 1 cm³ 10 % KJ and titrated with $n/1000$ $Na_2S_2O_3$ with starch as indicator.

The solubility of $Pb(JO_3)_2$ in concentrated solution of KJO_3, unmeasurable with ordinary chemical methods, was determined by the method of radioactive indicators. A little, but precisely weighed quantity of $Pb(JO_3)_2$ was mixed with traces of RaD (isotope of Pb), measured in arbitrary units in an electroscope using the ionisation made by its decay product—RaE (isotope of Bi). A saturated solution of this salt at 25° was prepared and the samples of it were evaporated and measured in electroscope. As isotopes were unseparable, the ionisation made by the samples gave immediately what quantity of the salt was dissolved in them. This method gave the same result as the iodometric titration. For solubility determinations in presence of great excess of KJO_3 an electroscope was replaced by a more sensitive Geiger-Müller electronic counter. The samples of the saturated solutions of $Pb(JO_3)_2$ + RaD in presence of KJO_3 were treated with 5 cm³ 0.4 % $Pb(NO_3)_2$ and then with 5 cm³ 1 %

KJO_3. The precipitated $Pb(JO_3)_2$ + RaD was filtered off and measured with the help of the counter. The solubility was calculated by the comparison of the radioactivity of the sample with the radioactivity of a sample of the solution in water (where the solubility was known), treated in the same way

Results. The solubility of $Pb(JO_3)_2$ in water at different tem-

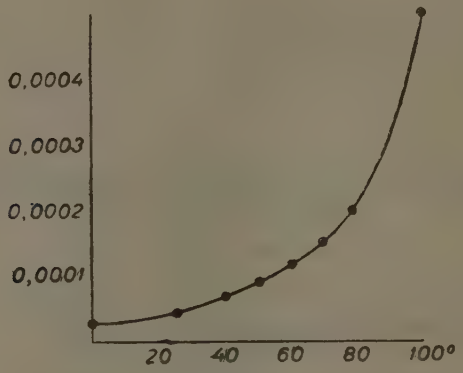

Fig. 1.

peratures is given in Table 1. and in Fig. 1, where for comparison the result by Harkins and Winninghoff at 25° is also given.

Table 1

Solubility of $Pb(JO_3)_2$ in water

t°	mg/l	mol/l
0	15.04	0.0000270
25	29.25	0.0000525
40	44.3	0.0000795
50	52.0	0.0000934
60	61.0	0.000109
70	79.5	0.000143
78	114	0.000205
100	275	0.000495

The solubility of $Pb(JO_3)_2$ in concentrated solutions of KNO_3, $Ca(NO_3)_2$ and KJO_3 at 25° is given in Table 2 and in Fig. 2, where the solubility of $Pb(JO_3)_2$ is plotted against the total ionic strength of the solution. The extrapolation of this curve to zero ionic strength gives the limiting solubility of $Pb(JO_3)_2$. For 25° it is 29.22 mg/l. The activity coefficients are given in the last column.

The dotted curve in Fig. 2 gives the activity coefficients against the ionic strength.

Table 2

Solubility of $Pb(JO_3)_2$ in concentrated solutions of electrolytes at 25°

Electrolyte	Concentration mol/l	Solubility		Total ionic strength	Activity coefficient
		mg/l	mol/l		
—	—	29.25	0.00005255	0.000315	0.999
KNO_3	0.2	58.5	0.0001051	0.4006	0.500
»	1.4	186	0.000333	2.802	0.157
»	2.25	307	0.000551	4.503	0.0953
$Ca(NO_3)_2$	1.30	362	0.000651	7.704	0.0807
»	2.27	585	0.001050	13.625	0.0500
»	3.07	803	0.00144	18.429	0.0364
»	3.6	929	0.00167	21.61	0.0315
»	4.22	1030	0.00185	25.33	0.0283
»	5.18	1100	0.00197	31.09	0.0267
KJO_3	0.0001	7.6	0.0000136	0.000282	1.003
»	0.001	0.3	0.00000053	0.0020032	0.932

Summary

1. The solubility of $Pb(JO_3)_2$ in water at temperatures from 0° till 100° was determined.

2. The solubility of $Pb(JO_3)_2$ in concentrated solutions of KNO_3 and $Ca(NO_3)$ at different concentrations was determined

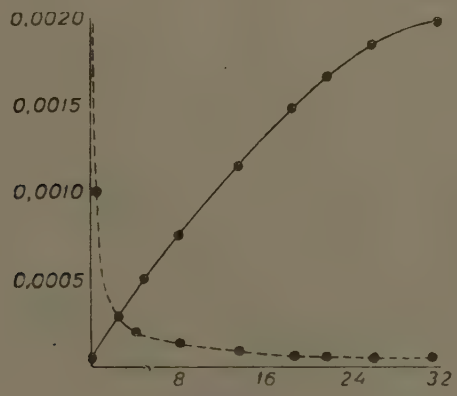

Fig. 2.

3. The solubility of $Pb(JO_3)_2$ in concentrated solutions of KJO_3 was determined by means of radioactive indicator RaD.

4. The limiting solubility and the activity coefficients at different ionic strength were calculated.

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CHEMISTRY

CASES OF MOBILITY OF THE NITRO-GROUP

By K. S. TOPČIJEV

(*Communicated by A. E. Favorskij. Member of the Academy, 13. XI. 1935*)

NITRO-GROUP MOBILITY IN 3, 4-DINITROANISOL

The theoretical interest afforded by a detailed study of cases of mobility of the nitro-group is enhanced by the fact that these reactions, in as far as they affect fatty and, what is more interesting still, heterocyclic compounds, are entirely unknown. The little that is known in this respect with regard to aromatic compounds strikes one as being entirely unadequate as compared with the importance of the problem [1].

This problem has thus been but very insufficiently studied, not only in its theoretical, but also in its practical aspect.

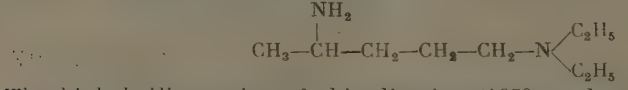
Such references to the mobility of the ortho-and para-nitro-groups as do occasionally occur are but formal statements, and, owing to the equivalence of the position, they can be used in solving the important question of as to which of the nitro-groups is the mobile one, only in the case of pure *o*- and *p*-dinitro-derivatives devoid of any other substitutes in the benzene nucleus. These statements thus refer to an exceptional case. In the complex cases of polynitro-compounds, especially if the latter also contain some other substitutes, the question as to which of the nitro-groups is the active one cannot at all be solved on the basis of these general statements, even if the compound in question had several ortho-para-positions.

This paper on the mobility of the nitro-group describes a new method devised by us for bringing about such a reaction in the case of 3, 4-dinitroanisol, which is of special interest to us. We hope to be albe to increase by further investigations the available evidence with reference to the heterocyclic compounds.

All we shall say with regard to the mechanism of the reaction and the methods of determining the position of the mobile nitro-group in the molecule of the reacting substance is that the existing electronic conceptions, irrespective of their relative value for the region in question, have so far been the only method that enabled us tentatively to determine the position of the mobile nitro-group in the molecule of substances introduced into the reaction of substitution. As further evidence becomes available, it will be possible to state in how far one would be justified in using these conceptions to interpret the phenomena connected with such facts of the mobility of the nitro-group as have lately come to our notice.

In selecting the substances to be substituted for one of the nitro-groups in 3, 4-dinitroanisol, we were guided, in addition to the interest afforded by the products of the reaction, first, by the facilities for studying the reaction within a wide range of temperatures, and, secondly, by the possibility of obtaining unimpeachable evidence of the structure of the product to be obtained.

We therefore chose for our experiments the reaction of 3, 4-dinitroanisol with 1-diethylamino-4-aminopentane



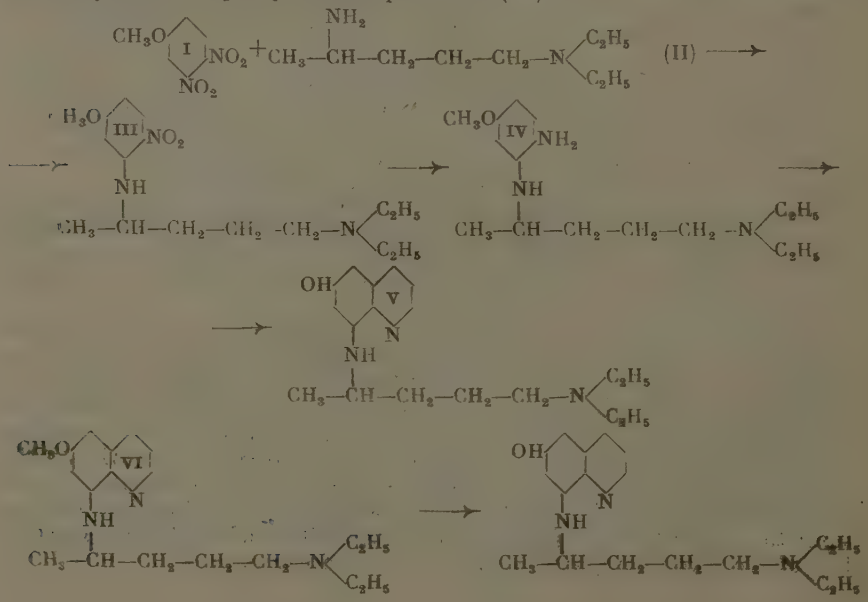
The high boiling point of this diamine (195°) makes it possible to bring about the reaction by simply heating the reacting substances at high temperatures without pressure.

Another possibility of studying the structure of the products obtained is afforded by Skraup's synthesis of the well-known (2) derivatives of 8-amino-quinoline from the appropriate amines.

The primary amino-group forming part of 1-diethylamine-4-aminopentane is substituted for the halogen in molecules possessed of a mobile halogen.

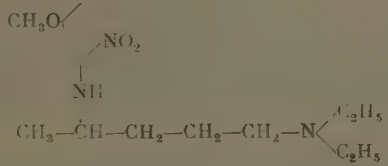
The amino-group of 1-diethylamino-4-aminopentane appears to react with equal facility with the nitro-group of 3,4-dinitroanisol (I), which is in position 3, this having been proved both by the reduction of the nitro-compound (III) produced by the reaction, and by Skraup's treatment of the fatty aromatic amine (IV).

Simultaneous with Skraup's reaction is the saponification of the methoxyl group, the 6-oxy-8-diethylamine-isopentane-amino-quinoline (V) separated as a result of the reaction being in all points identical with the saponification product of methoxyl in the well known 6-methoxy--8-diethylamino-isopentyl-amino-quinoline (VI):



EXPERIMENTAL

1. Reaction between 3,4-Dinitroanisol and 1-Diethylamino-4-Aminopentane. 15 g of pure 3,4-dinitroanisol, melting point 70°, ground to a fine powder, are mixed with 15 g of pure 1-diethylamino-4-aminopentane, boiling point 195°, in a flask provided with a vertical tube. In the process of mixing, the anisol is dissolved in the amine. The flask is carefully heated to 75—80°, a sudden and violent reaction setting in during heating. At that moment the source of heat is removed, and the contents of the flask allowed somewhat to settle. Heating is then resumed, the temperature of the oil bath being gradually brought up to 140—150°. At this temperature the reaction is continued for about 8 hours, an evolution of buff-coloured nitrogen oxides through the condenser tube being observable throughout the reaction. Thereafter, as soon as a piece of Congo paper wetted in water is no longer stained blue when placed at the upper end of the condenser tube, the heating is stopped. The flask is then connected to a vacuum, and the excess of the diamine is distilled off through the cooler, about 4 g being thus distilled off. The residue is treated with 30% acetic acid to remove the non-reacting portion of the dinitroanisol. The tar is then filtered off the solution obtained. The acid filtrate is treated with benzene, which absorbs the non-reacting portion of the dinitroanisol. After separation from the benzene, the lowermost aqueous layer is treated with strong alkali and 5 times extracted with 30 cm³ of ether. After drying the combined ether extracts with potash and the distillation of ether, there remains about 20 g of a heavy yellow oil, which stains the skin an intense yellow. This oil was distilled *in vacuo*. Boiling point 205° at 2 mm.



3-[(1-diethylamino-4-isopentyl)-amino] -4-nitroanisol.

	Analyses		Calculated %		Found %	
Weight of sample used	CO_2	H_2O	C	H	C	H
1) 0.1230	0.2803	0.1016	62.13	8.73	62.15	9.24
2) 0.1335	0.3060	0.1094	62.13	8.73	62.51	9.17

2. Reduction of 3-Diethylamino-Isopentyl-Amino-4-Nitroanisol. 20 g of the nitrogen compound was dissolved in 70 cm³ of concentrated hydrochloric acid, 65 g of stannous chloride and 100 cm³ of concentrated hydrochloric acid being slowly added to the solution.

The solution was allowed to stand for 2 hours, whereupon a strong solution of caustic soda was added to it until the stannic hydrates separated had been completely dissolved. The mixture was repeatedly extracted with ether. The ether extracts were dried with potash, the ether being then distilled off. The oily residue was distilled *in vacuo*. The 3-(diethylamino-isopentyl-amino)-4-aminoanisol thus obtained (14 g) is a colourless oil, which becomes readily oxidized on exposure to air. Boiling point 190—192° at 3 mm.

3. Skraup's Synthesis of 3-(Diethyl-amino-Isopentyl-Amino)-4-Aminoanisol. 7 g of amine, 4.5 g of arsenic acid-9.0 g of glycerin and 15 g of sulphuric acid were taken; the whole was carefully mixed and heated in the usual manner over an oil bath for 6 hours at 170°.

At first the contents of the flask had the usual appearance, characteristic of Skraup's synthesis, but soon the mass began to froth, and towards the end of the reaction the frothing increased considerably.

The contents of the flask was diluted with water to a volume of 250 cm^3 and allowed to stand overnight. The next day, the liquid was filtered off, a residue of buff colour being left behind. The filtrate was boiled for a short time with animal charcoal and then filtered off. A solution of alkali was added to the filtrate after cooling, the whole, however, remaining in solution. A strong current of carbon dioxyde was then passed throuhg the solution, causing the precipitation of an insoluble residue of 6-oxy-8-diethyl-amino-isopentyl-aminoquinoline, which soon assumed an oily consistency. It was separated with acetic ether, distilled *in vacuo*, and proved identical in all points, including boiling point (210—215°) at 2 mm, with the 6-oxy--8-diethylamino-isopentyl-aminoquinoline obtained by a 6-hours heating of 6-methoxy-8-diethylamino-isopentyl-aminoquinoline in a sealed up tube at 150°, with concentrated hydrochloric acid. The oxy-compound obtained is identical (in respect of constants) with the product described in the Swiss patent of IGF and obtained by the saponification of the methoxyl of an analogous methoxy-compound with hydrobromic acid.

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ON THE PREPARATION OF 3, 4-DINITROANISOL

The fact of the mobility of one of the nitro-groups in 3, 4-dinitroanisol, described above, had necessitated, for its detailed investigation, the use of considerable quantities of dinitroanisol of the structure described.

Up to the present, only one method of the production of 3, 4-dinitroanisol has been described, viz. the nitration of meta-nitrophenol, the isolation of isomeric dinitrophenols obtained, and the methylation of the appropriate isomer ([1]).

An experimental verification of this method has shown that at all the stages of the process only poor yields are obtainable, the nitration of meta-nitrophenol, which in itself is a rather rare product, giving in a pure state only as much as 10—15% of the requisite 3, 4-isomer.

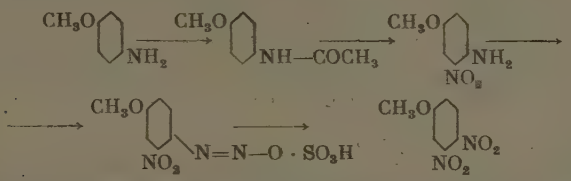
On the other hand, no 3, 4-derivatives are yielded by the direct nitration of either para-nitrophenol or para-nitroanisol, since the second nitro-group enters into the ortho-position with regard to the phenol or the methoxyl group ([2]).

A new method of synthesizing 3, 4-dinitroanisol is described here. The initial material used was para-anisidine. Acetylated para-anisidine is known

to give, upon nitration and saponification, a good yield of 3-nitro-4-aminoanisol (3).

The product obtained was diazotized, the nitro-group being substituted for the diazo-group.

The process follows the course given below:



The catalyst used in the last reaction of substitution (that of the nitro-group for the diazo-group) was the so-called Chevreuil salt, with which Hantsch used to obtain good results in reactions of substituton of the nitro-group for the diazo-group in some diazotized amines (4).

In our case, however, the methods employed by Hantsch were inapplicable owing to some complications, which proved so essential as to necessitate new procedures not described by Hantsch.

In using this catalyst, Hantsch applied it as a double compound of Chevreuil's copper salt and sodium nitrite, and introduced into the reaction of substitution the diazotized amine salt in a pure state, without the addition of acid. In our case, however, the excess of mineral acid used in diazotizing the amine destroys the catalyst.

Since the preparation of a solid salt of diazonium proved difficult in this case, while no diazotization could be brought about with the calculated amount of acid, as it gave rise to a combination of a part of the diazonium salt with the amine that had not yet been diazotized, the method had to be altered.

In order to prevent the formation of a diazo-aminocompound in diazotizing 3-nitro-4-aminoanisol, the latter was dissolved in a considerable excess of sulphuric acid, whereafter it was diazotized in the usual manner with a nitrite solution.

To the solution of diazonium so obtained was added some barium carbonate until the elimination of the acid reaction with Congo paper

When a solution of pure diazonium is added to the nitrite solution with the catalyst, a large volume of gas is evolved and a substance is precipitated, which appears to be a complex copper compound of the dinitro-product and the catalyst. On breaking up this complex with dilute hydrochloric acid, the whole of the copper is taken up by the solution, while the dinitroanisol becomes precipitated as a red powder. Recrystallized from alcohol, it melts at 71°, and is identical in all points, including melting point and the absence of depression in a mixed sample, with the methyl ether of 3, 4-dinitrophenol obtained by Bantlin by methylating 3, 4-dinitrophenol (5).

The yiled is 65 % of the theoretical.

EXPERIMENTAL

1. Diazotization of 3-Nitro-4-Aminoanisol. 35 g of finely ground 3-nitro-4-aminoanisol is placed in a flask containing 150 cm³ of water and 10 cm³ of sulphuric acid. The contents of the flask is heated over a water bath to 70°, and 20 cm³ of sulphuric acid is added. Then, while heating is continued, 100 cm³ or so of hot water is gradually added, until

nearly all has been taken up by the solution. The hot solution is quickly poured, while stirring, into a wide-necked vessel containing about 300 g of finely ground ice. The sulphate of nitroamine then separates as a very fine powder. The mixture obtained is cooled to +2° and diazotized at 2—3° in the usual manner with a solution of 14 g sodium nitrite in 30 cm^3 of water with continuous stirring. In the process, all is taken up by the solution, except a trifling amount of the diazoamino-compound. The solution is filtered, and the filtrate treated with finely powdered barium carbonate until no more carbonic acid evolves and no acid reaction with Congo paper is noticeable. The solution is filtered, and the clear filtrate is introduced into the reaction of substitution.

2. Preparation of Copper Catalyst. A concentrated solution of copper sulphate is precipitated with a solution of caustic soda, and the precipitated hydrate is filtered in a Büchner funnel; the residue is lightly rinsed with water, drained and transferred into a flask while still moist. Here it is stirred into a homogeneous mass with an equal volume of water, a strong current of SO_2 being then passed through the flask while stirring vigorously. While this is being done, the hydrate is gradually taken up by the solution, dying the latter emerald green. The flask is placed on a wire net and heated to boiling, the current of SO_2 continuing to circulate through it. After the precipitation of a bright-red crystalline residue has begun, the flask is heated for another 15 min., the SO_2 current being still maintained. Upon cooling, the precipitate is filtered, washed with water and dried in the air. The salt so obtained is a bright-red crystalline powder. The composition attributed to this salt by Rammelsberg is: $CuSO_3.Cu_2SO_3 \cdot 2H_2O$ [6].

3. Substitution of the Nitro-group for the Amino-group in 3-Nitro-4-Aminoanisol. 40 g of cupric salt is slowly added with stirring to a solution of 120 g sodium nitrite in 200 cm^3 of water. After the disappearance of the reddish tint of the solution due to the addition of the copper salt, a solution of diazotized amine is gradually added, with careful stirring. The addition of every portion is accompanied by the evolution of gas and by the simultaneous precipitation of a dirty-grey residue. When the whole of the solution has been poured in, the stirring is continued for another hour, and the precipitate drained. It is washed with water, placed in a flask while still moist, shaken with an equal volume of water, and some strong hydrochloric acid is added to it. In the process, the whole of the copper is taken up by the solution, and simultaneously a red crystalline residue of 3,4-dinitroanisol is precipitated. It is drained, rinsed in water and dried in the air. About 25 g is obtained. Recrystallized out of alcohol, it assumes the shape of golden needles and melts at 70—71°. A sample mixed with the 3,4-dinitroanisol obtained by methylating 3,4-dinitrophenol melts at 70°. 3,4-dinitroanisol may be distilled *in vacuo* without decomposition; in that case its boiling point is 168 at 3 mm.

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MINÉRALOGIE

DÉTERMINATION AU MICROSCOPE DES INDICES DE RÉFRACTION DES SUBSTANCES RÉSINEUSES ET DES MINÉRAUX DE HAUTE RÉFRINGENCE

Par N. VEDENEJEVA, S. GRUM-GRŽIMAJLO et A. VOLKOV

(*Présenté par S. I. Wawilow, de l' Académie, le 18. VII. 1935*)

Pour accroître la précision et la sûreté de la détermination des indices de réfraction des minéraux de haute réfringence par la méthode d'immersion, et pour éviter, d'autre part, la fâcheuse nécessité de contrôler les indices de réfraction des milieux d'immersion avec des prismes au goniomètre, nous avons eu recours à la méthode interférentielle, telle qu'elle a été proposée par le professeur A. A. Lebedev (1) pour le microscope polarisant. L'interféromètre a été exécuté d'après les dessins mis à notre disposition par l'Institut d'Optique. Nous avons étudié en qualité de milieux d'immersion de haute réfringence des alliages de pipérine avec les iodures de As et de Sb, des mélanges de soufre amorphe avec le sélénium et des mélanges isomorphes de cristaux isotropes du bromure et du iodure de thallium recommandés par T. Barth.

Nous nous servions d'abord d'une lunule sphérique dans le verre, que l'auteur de la méthode et le professeur Zavarickij pratiquent pour la détermination des indices de réfraction des liquides. Mais nous avons constaté ensuite que, pour diverses raisons, cette lunule ne nous convenait pas. Dans le cas, par exemple, des mélanges de soufre avec le sélénium, nécessitant un échauffement considérable suivi d'un refroidissement brusque de la préparation, ainsi que dans le cas des mélanges de TlBr + TlI, dont le point de fusion est voisin de 400°, la lunule peut facilement se fendiller ou bien devenir biréfringente.

Il convient d'y ajouter que la plupart des milieux d'immersion dont il s'agit absorbent fortement la lumière, tandis que la lame de la préparation garnissant la lunule est trop épaisse et souvent opaque. Le procédé même du coulage de la préparation dans la lunule est minutieux, et les bulles d'air qui se forment facilement entravent les mesures.

Pour tourner toutes ces difficultés nous avons procédé de la façon suivante: au lieu du précédent arrangement nous prenons une lentille plan-convexe et l'appliquons sur la préparation avec la surface convexe, en observant bien que la surface plane soit parallèle à la lame de verre qui supporte la préparation. La lentille fournit le même tableau interférentiel que la lunule, c'est-à-dire une suite de franges équidistantes.

Nous avons construit un petit fourneau-presse servant à fondre la préparation et à faire coller la lentille de telle façon que sa surface reste horizontale. Une pincette spéciale permet au besoin, en opérant, par exemple, avec des alliages, de saisir rapidement la lentille avec la préparation échaufée qui y adhère et deposer sur l'eau la lame de verre supportant la préparation. En procédant ainsi on ne risque pas d'abîmer la lentille, puisque pratiquement elle n'a pas le temps de s'échauffer.

L'exactitude de la détermination au moyen de la lentille dépend grandement (tout comme dans la cas précédent, d'ailleurs) de la différence $N—N_1$ des indices de réfraction du verre et de l'objet.

La dite différence $N—N_1$ est liée à la différence de marche mesurée et à la distance séparant deux franges d'interférence voisines par les formules $N—N_1=k\delta=c/s$, où k et c sont des constantes dépendant de la courbure et des dimensions de la lentille et du déplacement l des rayons interférents. Dans nos expériences nous avons préféré de mesurer la distance entre les franges, vu que ce procédé est techniquement plus simple et plus sûr, en tant que le résultat ne dépend pas des erreurs de graduation du compensateur. La constante c a la valeur $c=(\lambda r)/(la)$, où λ désigne la longueur d'onde, l—le déplacement des rayons interférents, r—le rayon de la lentille et a—la valeur de la division du micromètre oculaire.

L'expression de l'erreur du résultat:

$$d(N-N_1)=(N-N_1)\left(\frac{dc}{c}+\frac{ds}{s}\right) \qquad (1)$$

indique que la définition de c et de s exige une précision relative de plus en plus élevée à mesure de l'accroissement de la différence $N—N_1$. Déjà en supposant $N—N_1=0.2$, nous devons avoir $dc/c+ds/s\geqslant 0.005$, s'il s'agit de mesurer N_1 à 0.001 près.

Pour obtenir ce degré d'approximation il est nécessaire: 1) que les lectures soient faites au micromètre en nombre suffisant, 2) que le plan dans lequel on observe le tableau d'interférence soit toujours minutieusement mis au foyer* et 3) que la constante c soit définie non pas indirectement par les grandeurs r, l et a, mais directement, en graduant la lentille à l'air, l'eau étant aussi d'un emploi avantageux à cet effet.

D'autres procédés de graduation de la lentille et de la lunule donnent des résultats bien pires, ce qui nous amena à les abandonner.

Nous avons déterminé la dispersion de 16 alliages de pipérine avec les iodures, en vue de les utiliser comme milieux d'immersion, en appliquant la méthode de variation chromatique. Ces alliages ont été préparés par le professeur A. A. Koĉetkov au Laboratoire de Chimie organique de l'Université de Moscou. La teneur en iodures augmente de 4% d'alliage à alliage (en commençant par 24% et jusqu'à 84%). Les mesures se faisaient à la lumière monochromatique pour des longueurs d'onde de 670, 589, 577.5 et 546 μμ.

Les traits verticaux sur les courbes 6, 11, 15 et 16 représentent la valeur de l'erreur calculée selon la formule (1); la longueur de ces traits croît à mesure de l'accroissement de la différence $N—N_1$. Elle prend dans quelque cas (courbe 11, $\lambda=546$ μμ) des valeurs excessivement élevées, grâce à quelques mesures des distances entre les franges d'interférence qui par hasard n'ont pas été effectuées assez soigneusement.

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* Ce plan se trouve près du sommet de la lentille; pour le fixer on trace au diamant deux petits traits de deux côtés du sommet. Ces derniers peuvent servir en même temps de repères pour centrer la frange d'interférence (en opérant avec le compensateur) dans les mesures des différences de marche en cet endroit de la lentille.

La mesure au moyen de la méthode interférentielle a confirmé la variation des indices avec le temps que nous avions constatée avec des prismes en alliages de pipérine (²) et qui avait déjà été indiquée auparavant par Larsen. Cette variation, ainsi que le fait d'une diminution sensible de l'indice à la suite d'un échauffement de l'alliage (cas qui peut toujours se présenter), crée la nécessité de contrôler constamment la grandeur N_1.

Il serait évidemment désirable d'effectuer ces mesures de contrôle sur la même préparation, à laquelle serait incorporée par fusion de la poudre du minéral soumis à l'examen. Cette question acquiert une importance particulière lorsqu'il s'agit d'alliages de soufre avec le sélénium, où, selon les quantités de λ- et μ-soufre qui se forment à la confection de la préparation, les variations de l'indice de réfraction peuvent atteindre jusqu'à des unités de la deuxième décimale.

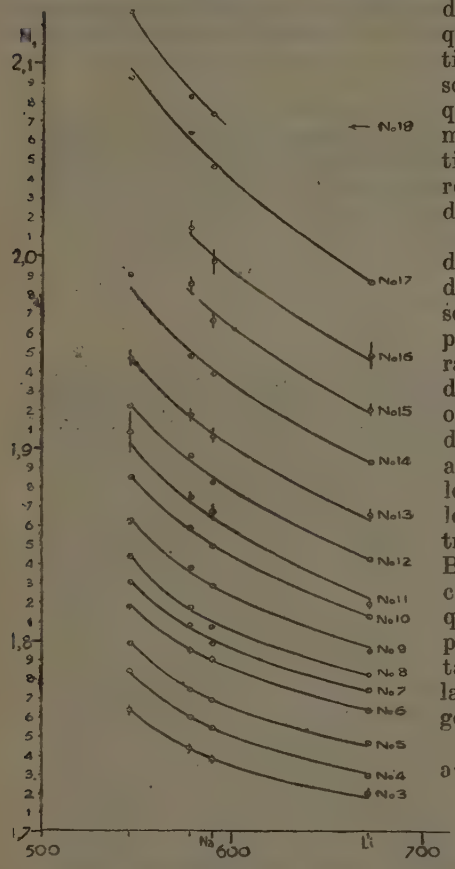

Fig. 1.

L'application de la lentille permet de résoudre facilement cette question, dont l'importance est manifeste non seulement pour le cas enivisagé, mais pour la méthode d'immersion en général, et de déterminer l'indice du milieu d'immersion au même endroit où l'on observe la ligne de Becke: on fait fondre sur la lame de verre la préparation avec la poudre à examiner et recouvre le tout avec la lentille. Mettant la lentille du côté plan en bas, on peut travailler en observant la ligne de Becke dans les conditions ordinaires, c'est-à-dire avec des grossissements quelconques. La petite quantité de poudre sous la lentille n'altère pas le tableau d'interférence et n'entrave pas la mesure de la distance entre les franges d'interférence.

On peut observer l'interférence avec la lentille même dans des cas aussi difficiles (alliages de sels de Tl) que ceux qui sont présentés par des conglomérations de cristaux isotropes. Mais alors, pour tenir compte de la haute température de fusion de l'objet à examiner, les lentilles doivent être faites en pyrex optique ou en verre de quartz, d'où résultent des conditions défavorables pour la précision de la mesure ($N—N_1$ étant plus grand que 1). Ici donc, la méthode interférentielle exige encore un perfectionnement subsidiaire.

L'application de la lentille permet de résoudre une série de questions de réfractométrie pratique. La lentille est, par exemple, d'emploi commode pour des substances telles que résines, huiles desséchantes, vernis, mastes

plastiques transparentes. L'experience montre qu'elle peut être utilisée pour l'examen de substances possédant un grand pouvoir d'absorption, telles que les asphaltites etc.

En cassant et enlevant un coin de la lame de verre couvrant l'objet microscopique (lames minces, par exemple) et si l'on met la lentille légèrement échauffée sur la couche ainsi mise à découvert de baume du Canada, il est facile de déterminer l'indice de réfraction de ce dernier.

Nous étudions actuellement le baume de sapin blanc. L'examen effectué d'abord sur un seul échantillon montre que ces deux baumes se touchent de bien près, tant pour l'indice de réfraction que pour le coefficient de température.

Notons, enfin, qu'au lieu de la lentille nous nous servions parfois d'un morceau de baguette de verre bien polie de forme cylindrique régulière, tranchée suivant l'axe, sur lequel nous faisions au moyen d'un diamant deux courtes entailles pour fixer l'endroit où se fait la mesure.

Cabinet d'Optique des Cristaux
de l'Institut des Matières Premières Minérales.
Moscou.

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BIOLOGIE

ÜBER DIE ENTWICKLUNGSQUELLEN DES LATERO-VENTRALEN TEILES DER RUMPFMUSKELN SOWIE DER KAUDALEN MUSKULATUR BEI AMPHIBIEN

Von Z. KATZNELSON

(Vorgelegt von A. A. Richter, Mitglied der Akademie, d. 10. VI. 1935)

In den vorigen Arbeiten ([1],[2]) ist von mir der Nachweis erbracht worden, dass bei den Amphibien die Myotome nicht die einzige Quelle der Entwicklung der quergestreiften Muskeln sind. Meine Beobachtungen haben die Befunde einer Reihe von Verfassern ([3],[7],[8],[9]), bezüglich der Möglichkeit der Entwicklung der quergestreiften Muskulatur durch lokale Differenzierung der Mesenchymelemente, bestätigt. Lewis konstatierte die mesenchymale Histogenese der Muskeln im Laufe seiner Erforschung der Entwicklung des Armes des menschlichen Embryos, Wassjutotschkin—indem er die Thymusdrüse des Huhnes untersuchte. Schmidt erforschte die mesenchymale Myogenese in der Zunge und den Gliedmassen verschiedener Säuger.* Ich habe an Urodelen eine mesenchymale Anlage der quergestreiften Muskeln für die Muskeln der Gliedmassen, der Kiemen, der ventralen Wand des Abdomens und für einige Kranialmuskeln festgestellt.

Indem ich eine Neuuntersuchung der Myotomalmuskulatur (der Rumpf und Kaudalmuskeln) unternahm (für deren Notwendigkeit ich in den erwähnten Abhandlungen den Nachweis erbracht habe) konstatierte ich, dass auch hier das Mesenchym in gewissem Masse an den histogenetischen Prozessen mitbeteiligt ist. Diese Mitbeteiligung besteht nicht allein darin, dass, wie dies gewöhnlicherweise angenommen wird, auf Kosten des Mesenchyms Elemente des Endomysiums entstehen; meine Beobachtungen zeigen, dass zu den Myotomanlagen Mesenchymelemente sich hinzugesellen die im Sinne der Ausbildung von quergestreiften Muskelfasern sich differenzieren.

Als Untersuchungsobjekt dienten mir, wie früher, Larven der sogen. Ural Salamander (*Salamandrella keyserlingi* Dyb.) und des Axolotls.

In Betreff der Rumpfmuskulatur ergibt sich aus meinen Beobachtungen Folgendes. Der dorsale und dorso-laterale Teil der Rumpfmuskulatur entsteht gänzlich aus Myotomen. Von den letzteren entspringt zwischen dem Ektoderm und den Derivaten des Mesoderms der sogen., von Maurer beschriebene ([4],[5]) Myotomfortsatz, wobei der genannte Verfasser meint, dass der gesamte

* Näheres darüber siehe in meiner Schrifttenübersicht.

laterale wie ventrale Teil der Abdominalmuskeln eben auf Kosten dieses Fortsatzes sich entwickelt. Es ist bereits früher von mir erwähnt worden, dass der ventrale Teil der Abdominalmuskeln auf Kosten des Mesenchyms entsteht.

Meine neuen Beobachtungen zeigen, dass beim Studieren der Sagittal- und Querschnitte ein Anschluss typischer Mesenchymelemente an die Myotomanlagen zu sehn ist. Der myotomale Ventralvortsatz bildet wohl nur die Basis für die Anlage der abdominalen ventralen (latero-ventralen) Muskeln, während im weiteren die Myogenese auf Kosten des Mesenchyms vor sich geht. Im typischen Mesenhymsyncytium gelingt es quergestreifte Myofibrillen und Gestaltung der primären Muskelfasern aufzudecken. Ich habe niemals eine Differenzierung von Myofibrillen in isolierten Mesenchymzellen zu sehen bekommen [wogegen A. M. Wassjutotschkin (9), dieses in der Histogenese der myoiden Elemente der Thymusdrüse beobachtete]; aus dem losen Mesenchymreticulum entsteht immer ein Symplast, dessen Protoplasma einen ektoplasmatischen Charakter (im Sinne Studnicka's) erlangt; daraufhin beginnt in einer derartigen symplastischen* mesenchymalen Anlage die Differenzierung der Myofibrillen. Bei 26—32 Tage alten Larven lässt sich zwischen den Elementen der Reihe: Mesenchymzellen—Mesenchymsyncytium—Mesenchymsymplast mit in Bildung begriffenen quergestreiften Myofibrillen, eine Gamme gänzlich unmerklicher Übergänge aufstellen. Die auf der Grundlage des Mesenchyms entstandenen quergestreiften Fasern sind in nichts von den Fasern myotomaler Herkunft unterschieden.

Auch im Schwanze, geht, nach Abschluss der Differenzierung der Somiten, die Entwicklung der Muskulatur ebenfalls aus dem Mesenchym einher. Am Ende der zweiten und im Anfang der dritten Woche der Entwicklung lässt sich bei den Larven die volle Serie der Übergänge von den Mesenchymelementen an bis zu ausgebildeten Muskelsegmenten beobachten. Die Kaudalsegmente bestehen aus Ansammlungen von Mesenchymzellen, die zu einem Symplast zusammenfliessen und an bestimmten Stellen unter der Epidermis zu liegen kommen. In den abgelegendsten Kaudalsegmenten, die in dieser Weise angelegt werden, finden wir noch keine Myofibrillen vor. Kranialwärts bekommen wir Segmente mit einzelnen Myofibrillen, dann solche mit in Bildung begriffenen primären Muskelfasern und schliesslich ausgebildete, aus quergestreiften Muskelbündeln bestehende Segmente zu sehen.

An der Hand meiner Befunde halte ich es für möglich folgende preliminäre Schlüsse zu ziehen:

1. An der Entwicklung der Rumpf- und Kaudalmuskeln sind bei Amphibien Mesenchymelemente mitbeteiligt.

2. Der myotomale ventrale Fortsatz ergibt nur die Basis für die Anlage der Abdominalmuskulatur, zu der sich noch Muskelschichten hinzugesellen, die auf Kosten der zu dieser Anlage sich anschliessenden Gebiete des Mesenchymsyncytiums gebildet werden.

3. Auf Kosten des Mesenchyms wird der ventrale und ventro-laterale Teil der Rumpfmuskulatur, sowie die Muskelsegmente des Schwanzes gebildet.

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* «Syncytialon», laut der Terminologie Studnicka's.

ZITIERTE LITERATUR

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BIOCHEMISTRY

FLAVINS AND METABOLISM

I. FLAVINS AND AMYLOLYSIS

By A. J. CHARITE

(Communicated by A. A. Richter, Member of the Academy, 29. IX. 1935)

1. The importance of the oxidation-reduction potential, and of the oxydation-reduction systems, for the course of different fermentative processes has, of late years, been shown by the researches of many investigators. Liepmann showed the action of the redox potential on the course of the Pasteur reaction, Grossmann and Waldschmidt-Leitz and their collaborators elucidated the part played by the reduced form of gluthatione in the fermentative decomposition of proteins. Pringsheim, Borchardt and Hupfer (1) proved the capacity of gluthatione and cystine to act as a complement of amylase, this action being specific, since dithioglycolic acid is non-active as regards amylase.

It was to be expected that, while studying the role of flavins in different fermentative processes, we should be confronted with the question of this oxydation-reductive system's significance for the phenomena of amylolysis. It was necessary to verify whether flavins would exert the same action as glutatione did in Pringsheim's experiments, or would prove absolutely inactive in this process.

As in our experiments with proteolysis and glycolysis (Charite and Khaustow; Charite, Neifach and Morozowa, 1935), we used pure preparations of lactoflavin, kindly sent us by R. Kuhn (for which we again tender him our thanks), and methanol extracts from the liver, containing some other substances besides flavin. We determined the progress of the amylosis by the glucose formed, according to the Willstätter and Schübel method (5). The particulars of the experiments in the separate series are given below.

II. The first series of experiments was made with the addition of flavins only. Two sets of test-tubes were used.

1st set—control. 15 ml 1% soluble starch+1 ml 1% NaCl+13 ml phosphate buffer (pH=6.4)+1 ml ferment (1% solution of diastase (Merck) or a 10% solution of saliva).

2nd set—with flavins. 15 ml starch+1 ml NaCl+11 ml buffer+1 ml 1% solution of diastase+2 ml aqueous solution of flavins (=100 or 50 γ).

Both sets were placed in a thermostat at a temperature of 37°, taken out after 15 min. and subjected to further manipulations, according to the procedure of the method. The results obtained are expressed in ml $n/10$ J_2, not calculated in the form of sugar.

Table 1

Amylolysis under Addition of Flavins (with Saccharose)

Nos	Conditions	Amount of glucose formed in ml $n/10$ J_2		Difference in ml $n/10$ J_2	Annotations
		Control	With flavins		
1	Flavins 100 γ; duration of amylolysis 15 min.	3.0	3.3	+ 3.3	Diastase
2	» 100 » » » 2 h.	3.0	3.8	+ 0.8	»
3	Flavins 50 γ; » » » 15 min.	4.25	4.1	— 0.15	»
4	» » » » » » »	4.25	4.2	— 0.05	»
5	» » » » » » »	4.25	4.3	+ 0.05	»
6	» » » » » 10 min.	3.75	4.25	+ 0.5	»
7	» » » » » 3 h.	4.3	4.4	+ 0.1	»
8	» » » » » 15 min.	3.15	2.9	— 0.25	Salivary amylase
9	» » » » » 15 min.	3.16	2.8	— 0.3	»

The data speak against an action of flavins on amylolysis.

We see, in some cases, a considerable intensification of amylolysis under the action of flavins, attaining to as much as 0.8 ml J_2 while in other cases flavins actually cause an inhibition of amylolysis. The nature of the ferment (vegetable diastase or salivary amylase) seems to play no decisive part. In the experiments with salivary amylase we may, perhaps, note a tendency to the diminution of the amount of glucose formed.

The duration of the action of flavins on the ferment seems to be of no importance. In our experiments we extended the duration of the action from 10 minutes to 3 hours, without obtaining any appreciable difference in our results.

Similar results were obtained with the substitution of flavin-containing methanol liver extract for pure flavins. Some of the experiments shown below will illustrate this fact.

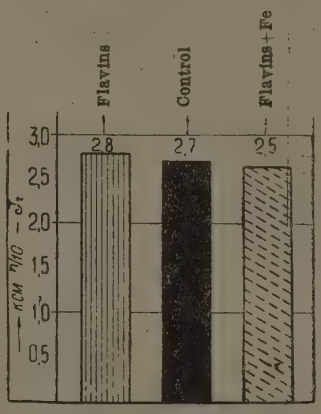
Table 2

Nos	Conditions	Amount of glucose formed in ml J_2 $n/10$		Difference in ml J_2 $n/10$	Annotations
		Control	With flavins		
1	Flavins 180 γ; 15 min.	2.45	2.2	— 0.25	Vegetable diastase
2	» 180 γ; 4 h.	2.9	2.8	— 0.1	»
3	» 110 γ; 15 min.	3.55	2.95	— 0.6	Salivary amylase
4	» 70 γ; 15 min.	2.2	2.0	— 0.2	»

Although the experiments with methanol extract resulted in a certain decrease of the glucose formed during amylolysis, yet it was within such narrow limits, that we ascribe it to the usual fluctuations of a biological

experiment, and not to the influence of the flavins. Still, here, too, we may observe, as the result of an interaction with salivary amylase, a somewhat more pronounced depression in the formation of sugar, which awaits its further elucidation.

III. The negative nature of the data obtained by us cannot yet be considered as definitively testifying to the absence of reciprocal action between flavins and amylolytic ferments. For the manifestation of this reciprocal action a preliminary activation of the ferment may possible be necessary. An activator of organic nature of the type of kinase, differing from Pringsheim's complement, was found by Waldschmidt-Leitz and Purr in the amylase system of the germinating seed. Aspartic acid according to Rockwood, in a 0.01 mol. solution, produces a 50% increase of amylolysis under the action of enzyme for the duration of an hour, and likewise protects the enzyme from decomposition.



Action of flavins on amylolysis

We repeated Rockwood's experiment with an addition of aspartic acid and flavins (methanol extract), but with no particular success.

We here append a description of the experiment:

Taken:

1) Control. 15 ml 1% starch solution+1 ml 1% NaCl+13 ml phosphates+1 ml 1% solution of diastase.

2) With aspartic acid+flavins. The same, but only 7 ml phosphate buffer and 5 ml flavins (=80%)+1 ml 0.15% aspartic acid.

3) Only flavins. The same, but without aspartic acid. In place of this, a supplementary 1 ml of buffer was taken. Time of incubation in thermostat—20 min.

Results obtained:

1) Control. Consumed J_2 (n/10): 3.45 ml
2) Flavins+aspartic acid (n/10): 3.3 ml
3) Flavins: 3.4 ml

These data prove that an addition of aspartic acid in the presence of enzyme + flavins does not activate these latter.

A combination of an organic complement with an inorganic salt sometimes activates the enzymatic process considerably. We see an example of this in the action of iron salts, conjointly with ascorbinic acid, on the enzymatic hydrolysis of gelatine (2). We built up a similar model from fravins with ferric and ferrous salts.

As representatives of the latter we used 1 ml n/10 $K_3Fe(CN)_6$ or 1 ml n/10 $K_4Fe(CN)_6$.

The procedure of the experiments resembled that of the tests made with an addition of aspartic acid, but instead of the latter we used 1 ml $K_3Fe(CN)_6$ or $K_4Fe(CN)_6$.

The data are summarized in Table 3.

As may be seen from the table, flavins even in conjunction with iron-salts, do not affect the course of amylolysis.

Table 3

Action of Iron Salts Added to the Enzyme + Flavins System

Nos	Conditions	Amount of glucose formed in ml J_2 n/10			Annotations
		Control	Flavins	Flavins+Fe	
1	Flavins 100 γ; 15 min.	3.5	3.6	3.55	1 ml $K_3Fe(CN)_6$
2	» » » »	3.95	3.7	3.85	» »
3	» » » »	3.15	3.8	3.0	1 ml $K_3Fe(CN)_6$
4	» » » »	2.2	2.35	2.1	» »
5	» » » »	2.65	2.5	2.25	» »

Discussion

In all the experiments performed by us with a view to elucidating the role of flavins in amylolysis, we obtained data that failed to prove an appreciable influence of flavins on this process.

The results of our experiments, however, may not be applied with full authenticity to the processes of amylolysis occurring within the animal cell. The procedure followed in our experiments was too much simplified as compared with the conditions existing in the cells. Warburg and Christian's experiments (4) on the action of a flavin-protein complex (the «yellow» ferment), on the decomposition of hexoso-phosphates is sufficiently instructive. A highly complicated system of ferment + co-ferment + Zwischenferment, besides a number of other favourable factors, is needed to ensure a corresponding effect. Therefore, to solve the problem we are confronted with, a series of further experiments is needed reproducing as closely as possible the conditions existing in the cell.

Our numerous experiments however, have left us the impression that an addition of flavins—especially under the action of salivary amylase—not infrequently leads to a decrease in the quantity of glucose formed.

Whether this is due to the action of flavins themselves or to that of some «accompanying» substances, it is as yet difficult to decide.

Summary

1. Lacto-flavin exerts no influence on diastase or salivary amylase outside of the cell.

2. The addition of iron salts or aspartic acid to amylolytical enzyme + flavin produces no appreciable action on it.

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BIOCHEMISTRY

FLAVINS AND METABOLISM

II. FLAVINS AND PROTEOLYSIS

By A. J. CHARITE and N. V. KHAUSTOV

(Communicated by A. A. Richter, Member of the Academy, 29. IX. 1935)

Waldschmidt-Leitz, Grassmann, and their collaborators have established the activating action of SH compounds on the proteolytic enzymes cathepsin and papain. In Bersin and Logemann's researches a conjecture is emitted concerning the non-specific nature of proteolysis activation by SH compounds. In their experiments papain was activated by succinic acid. According to their opinion, the activation of the enzyme consisted in the non-specific reduction, by succinic acid, of the prostatic groups of the enzyme. The investigations of Maschmann and Helmert (5) into the activating action of the vitamin C+Fe'' complex on papain and cathepsin, seemed to confirm this assumption. Purr (6), however, in his researches on the mechanism of proteolysis activation by an inactive preparation of papain, ascertained the activation of papain by SH compounds to be specific. The vitamin C+Fe'' complex and pyruvic acid+Fe'' were inactive in this case.

As to the activation of the proteolytic enzymes by the vitamin C+Fe'' complex, it could, in Purr's opinion, be explained, as well as Maschmann and Helmert's results, by the reduction of the SH compounds present in the preparations of papain and cathepsin. These latter would then activate the enzymes specifically. These redox systems would thus exert an indirect action.

Flavin is a redox system. The redox potential of this system is equal to —0.20 mV, i. e., it approximates the potential of the living cell. The value of the flavin potential shows flavin to be a mild oxidazing and a potent reducing agent. We therefore considered it necessary to verify the action of a flavin preparation as a reversible oxidation-reduction system of the processes of protein decomposition. We investigated the influence of flavins: 1) on the digestion of trypsin, 2) on the action of papain, and 3) on the autolytic process.

The Action of Flavin on Trypsin Digestion

Grassmann, Dyckerhoff and Shoenebeck (4) have established the activating influence of cystine, i. e. an oxidized form of SH, on trypsin. It was interest-

ing to ascertain whether flavin, too, would behave in the same fashion during the digestion of proteins by trypsin. The preparations of trypsin employed were ordinary market pancreatin and Merck's market trypsin. The flavin preparation contained an admixture of certain other substances and was obtained through the concentration of methanol exstracts from the liver, previously treated by chloroform (3). The activity of the trypsin was determined by Willstätter and Waldschmidt-Leitz's method. One of the experiments is given below.

1) 1 g pancreatin+2 ml buffer (pH=5.8)+5 ml albumin+5 ml water.
2) 1 g pancreatin+2 ml buffer+5 ml albumin+5 ml flavin solution (30 γ).

Table 1

	Quantity of ml 0.05 *n* KOH		
	At starting	In 18 hours	In 66 hours
1	0.9	12.8	17.3
2	0.9	12.8	17.3

The intervals between the activity determinations varied in the different experiments.

The influence of flavin on the action of trypsin may be expressed by the following curve (see Fig. 1.)

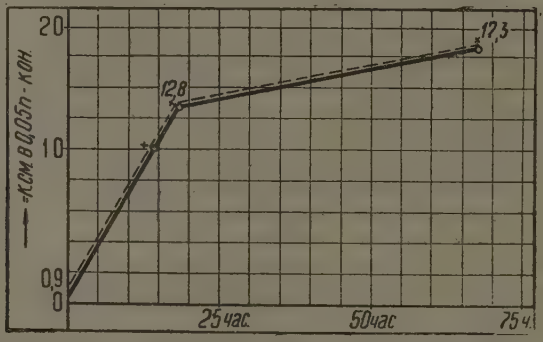

Fig. 1. The Effect of Flavins on the Digestion of Albumin by Trypsin: ××× without flavin; ○○○ with flavin.

К.СМ. В 0.05 n-KOH — cm³ in 0.05 *n* KOH; *час.* — hours

A comparison of the curves shows that flavin has no activating action on the digestion of albumin by trypsin. Neither does flavin retard this process.

Influence of Flavin on the Action of Papain

The flavin preparations used by us were: concentrated aqueous solutions of methanol concentrates from the liver, and a pure preparation of R. Kuhn's

lacto-flavin (we take this opportunity to express our deep thanks to Prof. Dr. R. Kuhn for the preparation of pure flavin which he so kindly set at our disposal).

The nature of the action exercised by the two flavin preparations on proteolysis was the same. The activity of the papain was determined by Maschmann and Helmert's method, and the degree of gelatin splitting, by van Slyke's. Experiments were made with free flavin and with flavin combined with Fe''. An example is given below of one of the experiments demonstrating the action of flavin concentrates prepared after our own method; also a table summarizing the results of some experiments.

1) 0.8 ml papain+2 ml buffer+12 ml 6.6% gelatine+5.2 ml water.
2) 0.8 ml papain+2 ml buffer+12 ml 6.6% gelatine+2 ml flavin (108γ)+1 ml M/100 $FeSO_4$+2.2 ml water.
3) 0.8 ml papain+2 ml buffer+12 ml 6.6% gelatine+2 ml flavin (108 γ)+3.2 ml water.
4) 0.8 ml papain+2 ml buffer+12 ml 6.6% gelatine+1 ml M/100 $FeSO_4$+4.2ml water.

Table 2

	At beginning	In 24 hours	Results (difference)
Control	2.1	4.7	2.6
Flavin + Fe'' . . .	3.3	5.2	1.9
Flavin	3.1	5.8	2.7
Control + Fe'' . . .	2.1	4.7	2.6

Table 3

Experiments with Kuhn's Preparations of Lacto-flavin

		Control	Flavin + Fe''	Flavin	Fe''
I	At starting	2.1	3.3	3.1	2.1
	In 24 hours	4.7	5.2	5.8	4.7
II	Difference	2.6	1.9	2.7	2.6
	At starting	4.2	3.1	3.5	3.0
	In 24 hours	5.9	4.9	5.7	4.7
III	Difference	1.7	1.8	2.2	1.7
	At starting	2.6	2.5	2.5	2.5
	In 24 hours	3.7	3.3	3.4	3.4
	Difference	1.1	0.8	0.9	0.9

The result of these experiments shows that neither flavin nor flavin+Fe'' have any activating effect on papain. The flavin employed for these tests was an aqueuos solution of methanol extracts from the liver. A question now arose as to whether these concentrates might not contain admixtures falsifying the true action of the flavins.

In order to settle this doubt, we had recourse to some further experiments with a pure crystalline flavin received from. R. Kuhn. The experiments

were conducted as shown in Table 4, but, instead of a flavin concentrate, 2 ml of an aqueous solution of flavins (100 γ) were added.

Table 4

Example of Experiment.

	At starting	In 24 hours	Difference
Control	2.6	3.7	0.9
Flavin + Fe″ . . .	2.5	3.3	0.8
Flavin	2.5	3.4	0.9
Control + Fe″ . . .	2.5	3.4	0.9

The results of the experiments may be represented by the following graph (see Fig. 2).

This figure clearly shows that neither free flavin nor flavin combined with Fe″ have any influence on the action of papain. This is probably due

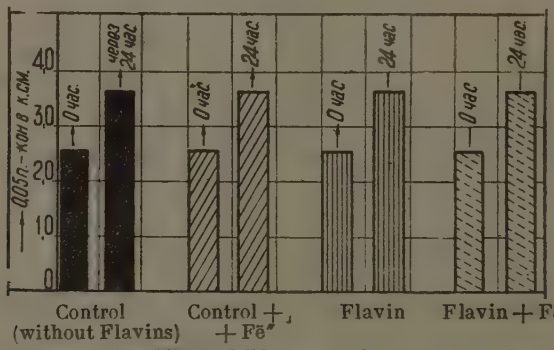

Fig. 2. The Effect of Flavin on the Papain Action.
0.05n.-KOH в K.CM—0.05*n* KOH in cm³
через 24 час.—in 24 hours; час.—hours.

to the fact that the experiments have been made under aerobic conditions. On the other hand, the flavin used for these experiments was in an oxidized state. And, finally, it might be presumed, that under conditions *in vivo*, i. e. in the presence of an entire enzyme complex, the flavin might, perhaps, have had a different effect. In order to verify the influence of flavin on the whole of the enzyme complex promoting proteolysis in muscular tissue, we resorted to experiments with autolysis of muscular tissue.

Influence of Flavin on Autolysis of Muscular Tissue

For this purpose we used the skeletal muscles of a rat. The experiments were conducted in two ways: 1) the muscle was crushed and suspended in a phosphate buffer (pH=5.0); toluene was then added as an antiseptic, and the muscle was autolysed in a thermostat at 38°. 2) The muscle was crushed

and suspended in 0.9% NaCl; toluene was then added, and the suspension autolysed in a thermostat at 38°. Flavin solutions were added to the experimental mixture in concentrations from 50 to 150 γ. The flavin was added either alone or combined with Fe''.

The increase in amino-groups was determined by van-Slyke's method. Experiments were conducted under aerobic conditions.

Experiment with Skeletal Muscle of a Rat

1) 5 g muscle+25 ml water+30.5 ml phosphate buffer.
2) 5 g muscle+17 ml water+8 ml flavin+30.5 ml phosphate buffer.

Table 5

Quantity of mg N_2

		In 24 hours	In 72 hours	Difference
1	1.15	1.1	2.40	1.39
2	1.09	0.96	2.54	1.58

An experiment with Kuhn's preparation was conducted in the same manner, but instead of a flavin concentrate an aqueous solution of crystalline flavin (100 γ) was added to the mixture.

Experiment with Heart Muscle of a Rat

1) 2.1 g muscle+21 ml 0.9% NaCl+0.5 ml toluene.
2) 2.1 g muscle+19 ml 0.9% NaCl+2ml flavin+0.5 ml toluene.

Table 6

Quantity of mg N_2

	At starting	In 24 hours
1	1.0	1.9
2	0.9	1.9

Experiment with Skeletal Muscle

1) 5 g muscle+50 ml 0.9% NaCl+1 ml toluene.
2) 5 g muscle+48 ml 0.9% NaCl+2 ml flavin+1 ml toluene.

Table 7

Quantity of mg N_2

	At starting	In 24 hours
1	1.0	1.6
2	1.0	1.6

The results of the experiments may be represented by the following graph (see Fig. 3).

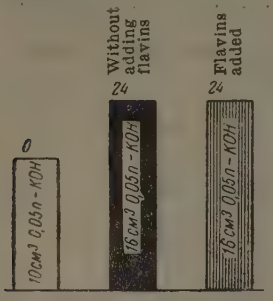

Fig. 3. Flavin influence on Autolysis of Muscular Tissue
0 час—0 hours; 24 час—24 hours

The data of the experiments conclusively show that neither liver flavin concentrates nor a pure preparation of flavin have any effect on the proteolytic enzyme complex of muscular tissue.

Discussion

Our work bears a preliminary character, and it is therefore difficult at present to interpret the data obtained without additional experiments. One of the possible reasons for this behaviour of the flavins might be the aerobic conditions under which the experiments were conducted. Further, in all the tests the flavin was used in its oxidized state. It is a well-known fact that the oxidized form of glutathione (GS—SG) is unable to activate papain. Should this analogy be extended to flavin, it would probably account in the same way for the results of the experiments.

As to the action of flavin on trypsin, the question is not yet clear. The oxidized form of flavin does not activate the action of trypsin, as opposed to the action of cystine. It might be possible here to emit a conjecture as to a specifically activating action of cystine on trypsin, whereas flavin is non-specific, as regards the mechanism of trypsin action and is therefore inactive. But in any case, this last question calls for further investigation.

The researches, begun by us, regarding the action of the leuco-form of flavin on proteolysis under anaerobic conditions, also the elucidation of the action of lumiflavin, may possibly give a definite answer to the question set by us in the beginning of this research, as to the nature of the influence of flavin on proteolysis.

Summary

The oxidized form of flavin under aerobic conditions does not activate the digestion of proteins by trypsin, does not activate the action of papain and does not activate the autolytic process of muscular tissue.

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GENETICS

ON THE FREQUENCY OF TRANSLOCATIONS IN DIFFERENT SECTIONS OF THE CHROMOSOMES IN *PISUM*

By S. I. KRAJEVOJ and R. A. RASSULY

(Communicated by A. A. Sapehin, Member of the Ukrainian Academy, 14. VIII. 1935)

In treating the seedings of *Pisum* with X-rays we unexpectedly discovered an interesting phenomenon consisting in a different frequency of cases of breakage in different sections of the chromosomes in relation to the spindle fibre. When we noticed indications of a regularity in the frequency of cases of breakage, we elaborated a method of recording the breaks in definite sections of the chromosomes. The method of determining the points where breakage occurs, is very simple and is based on direct observation of the breaks under the microscope. We prepared a standard corresponding to the scale of the chromosomes in microns. Then cases of chromosome breakage observed directly in the microscope were recorded under the corresponding scale division. The points of breakage can also be determined otherwise, for instance, by measuring the fragments. But this method was inappliable in our case, as we recorded the breaks of one chromosome only, and it is most difficult, if not impossible, to decide to what chromosome one or other fragment might belong. Chromosome 1 is 10 μ long, we divided the chromosome into sections (1 μ each) beginning with the place of constriction (which is also the place of attachment of the spindle fibre) outwards to its ends. For each dose we recorded the number of breaks observed directly under the microscope in a given number of nuclei (s. the table). The points of breakage were determined by means of a micrometer in accordance with the chromosome scale, and the number of cases of breakage was recorded under the corresponding division. The doses varied considerably, ranging from very weak ones (200 r) to lehtal ones (1200 r) for the given stage of the plant's development (s. the table). All the doses were applied at 130 kV max., 5 mA, 25 cm from the anticathode, with 1 mm aluminium filter. The apparatus used was the «Universal Transverter» of Koch and Sterzel.

Record of the dosing in relation to the points of breakage was carried out only for one pure line of *Pisum sativum vulgare* Alf., 010 BGSS. The results of these observations are given in the table. In the first column of the table the spindle fibre is marked zero, on both sides of it in ascending order are numbers showing the points of breakage in microns from the point of attachment of the spindle fibre. In the following columns of this table, the doses from 400 to 1000 r are shown, under the doses are the numbers of nuclei in which the breaks were counted, and below are the numbers of breaks corresponding to the dose and the point of breakage

Table

Scale of chromosome I in microns	Doses in r, number of nuclei examined and number of breaks			
	400/402	600/405	800/403	1000/400
3	0	0	0	3
2	5	7	11	14
1	11	13	23	33
0	0	0	0	0
1	17	23	41	48
2	15	21	37	29
3	9	13	19	25
4	2	5	7	14
5	0	1	3	7
6	0	0	0	0

The data of the table are represented in Fig. 1 as curves with the chromosome scale plotted along the axis of abscissae in microns. Point *C* signifies the place of attachment of the spindle fibre, and the figures to both sides of *C* signify the distance in microns from the place of attachment of the spindle fibre to the point of breakage (the short arm of the chromosome to the left, and the long one to the right). The absolute number of cases of breakage obesrved under the influence of X-rays was plotted along the, axis of ordinates. It should be noted that the method applied did not allow us to record the cases of breakage accompanied by mutual translocations.

It seems to us, however, that such cases cannot seriously affect the general trend of the regularity discussed below.

It is well known that the number of chromosome breaks increases with increased doses of X-rays. We examined 1610 nuclear plates of 133 seedlings.

The curves of Fig. 1 are approximately identical in their character, but quantitatively they all differ.

The curve (400 r) of the short arm (to the left of *C*) of chromosome 1, shows that 11 breaks were recorded at a distance of 1 μ from the place of attachment of the spindle fibre (see also the table), and that with an increase of the distance to two microns the number of breaks falls to 5. With increasing distance from the spindle fibre the number of breaks decreases more and more. The long arm of the investigated chromosome behaves similarly. 17 breaks were recorded at a distance of 1 μ from the place of attachment of the spindle fibre, 15 at a distance of 2 μ, 9 at a distance of 3 μ, and but 2 cases of breakage at a distance of 4 μ.

The curve for the dose of 600 r follows, in general, the same rule. Thus, in the short arm of the chromosome 13 breaks were recorded at a distance of 1 μ from the place of attachment of the spindle fibre (*C*), and but 7 cases of a distance of 2 μ. The long arm showed 23 cases of breakage at a distance of 1 μ from the constriction (spindle fibre), 13 cases at a distance of 3 μ, 5 cases at a distance of 4 μ, and but one case at a distance of 5 μ. The curve for the dose of 800 r has the same character but the figures are higher. 23 breaks were recorded at a distance of 1 μ from the constriction, 11 at a distance of 2 μ. The curve for the long arm with this dosage (800 r) shows 41 cases of breakage at a distance of 1 μ from the constriction, 37 cases at a distance of 2 μ, 19 cases at a distance of 3 μ, 7 breaks at a distance of 4 μ, and but 3 breaks at a distance of 5 μ. The dose of 1000 r gave the highest number of breaks.

33 cases of breakage were observed in the short arm at a distance of 1 μ from the point of attachment of the spindle fibre, 14 cases at a distance of 2 μ and but 3 cases at a distance of 3 μ. 48 cases of breakage were discovered at a distance of 1 μ from the spindle fibre of the long arm, 29 cases at a distance of 2 μ, 25 cases at a distance of 3 μ, 14 breaks at a distance of 4 μ, and but 7 breaks at a distance of 5 μ. This last curve showed in all the points except the section of the second micron a larger number of cases of breakage than the one for the dosage of 800 r. What is the most general and characteristic feature of all the above examined curves? The first thing to be noted is the existence of a zone of a maximum number of breaks at a distance of 1μ to both sides of the point of attachment of the spindle fibre (Fig. 1, *C*), this zone being followed by one of a lower frequency of

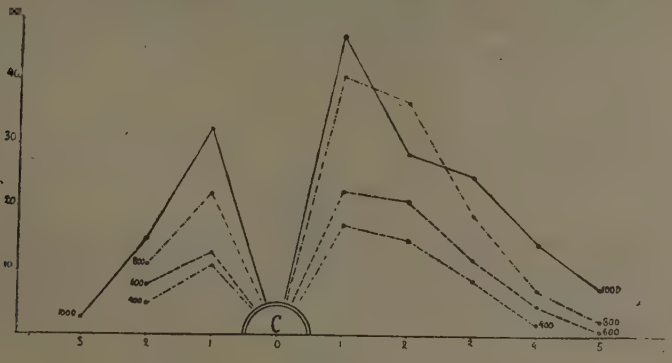

Fig. 1

cases of breakage etc. Thus, the number of breaks decreases with increasing distance from the place of attachment of the spindle fibre and at the ends of the chromosome falls to zero. Secondly, the number of breaks rises with an increase in the dosage, but no direct proportion between the dosage and the frequency of cases of breakage is to be observed. The decrease in the number of breaks with increasing distance from the point of attachment of the spindle fibre is more pronounced in the short arm of the chromosome (Fig. 1). It is characteristic of this curve that it is correlated with increased dosage, i. e. the curve shows a tendency to fall more steeply with an increase of dosage, as may be clearly seen in the curves for the short arm of the chromosome from point *C* leftwards. But the picture is somewhat different as regards the long arm of the chromosome. The number of breaks does not decrease quite so rapidly with increasing distance from the spindle fibre as in the short arm of the chromosome, especially in the case of two large doses (800 and 1000 r). As regards the long arm, the curve commencing from the point of the maximum number of breaks at a distance of 1 μ from the point of attachment of the spindle fibre shows a tendency to a more or less gentle slope, with the exception of the curve for the dosage of 1000 r, which sharply falls at a distance of 2 μ from the constriction. All the curves, both for the long and the short arms, overlie each other, only the curve for 800 r rises above the curve for 1000 r at a distance of 2 μ from the place of attachment of the spindle fibre (long arm of the chromosome), but perhaps this phenomenon is due to change.

The above facts lead us to the conclusion that points of chromosome breakage in *Pisum* under the action of X-rays are not contingent, and that the frequency of cases of breakage follows a certain regularity as regards their localization.

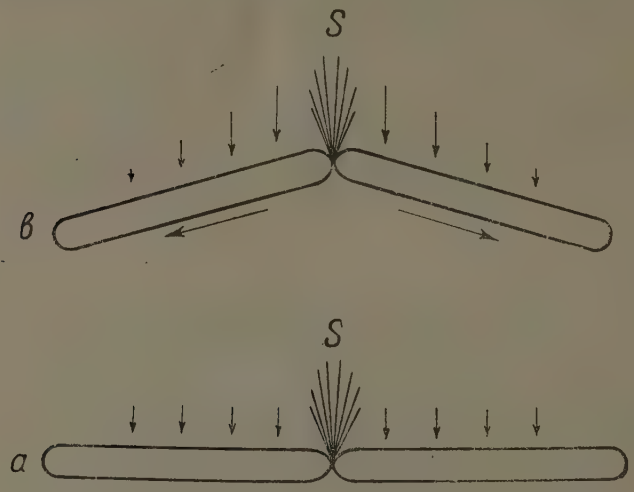

Fig. 2.

It follows from the above that the greatest frequency of cases of breakage in chromosome I of *Pisum* falls on the sections directly adjoining the place of attachment of the spindle fibre. With increasing distance from this point the number of cases of breakage gradually decreases and disappears entirely at the ends of the chromosome.

How should such a localization of breaks in different sections of the chromosomes be accounted for?

First Seckt ([5]) and then Williams ([6]) have established that there occurs an increase in plasmic currents on the irradiation of plant cells with X-rays, and that the viscosity of the plasm increases under certain conditions of irradiation. M. Zuelzer and E. Philipp ([8]), as well as Williams ([7]), have established the same under the action of radium rays on plant cells. By acting on plant organisms with X-rays and high temperature, De Mol ([3]) came to the conclusion that these factors modify the strength of the currents in the plasm and that they are probably due to a considerable extent to the phenomenon of the chromosome breakage. We shall make an attempt to reach an explanation of this phenomenon (localization of breaks) on the basis of the experimental data of the above-mentioned authors.

When the spindle fibre (*S*) is still in an akinetic state (Fig. 2*a*) the forces of the plasmic currents affecting the chromosome act uniformely along all its length (this is shown on the scheme by arrows of equal length). When the spindle fibre passes into the kinetic state (Fig. 2*b*) and, under changed conditions of currents and viscosity in the plasma, begins to draw the chromosome to the pole, the different sections of the chromosomes experience different resistances (Fig. 2*b*), the highest resistances being experienced by

sections adjacent to the spindle fibre (marked with the longest arrows in the scheme). This resistance decreases with increasing distance from the spindle fibre (Fig. 2, *b*, in the direction of the arrows). On the basis of these considerations, it is quite natural to suppose that the greatest number of breaks occurs in those sections where the surrounding plasm offers the greatest resistance. There exist in the literature some data that confirm the facts established above. By irradiating *T. Monococcum* with X-rays and by measuring the fragments, Câmara (1) has obtained data indicating that the regions of the chromosome nearest to the points of attachment of the spindle fibre are the critical regions of breakage. Glass (2) has found that the autosome regions in *Drosophila* lying near the spindle fibre are more liable to breakage than those removed from it. Patterson and his collaborators (4) have come to the conclusion that one maximum of breakage caused by the irradiation of the chromosomes of *Drosophila* lies in the region near the spindle fibre, while the other maximum of breakage lies near the ends of the chromosomes. No second maximum of breakage in *Pisum* has been observed by us.

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GENETICS

INHERITANCE OF DURATION OF VEGETATION PERIOD IN TOBACCO

By A. F. BUČINSKIJ

(Communicated by N. I. Vavilov, Member of the Academy, 8. VIII. 1935)

During the process of selective and genetic work with tobacco, the selection department of the All-Union Institute of Tobacco and Makhorka Industry has obtained very valuable material on the genetics of the separate characters in tobacco. This material justifies some generalizing conclusions about the inheritance of economically valuable characters in this plant, including the inheritance of the duration of the vegetation period.

This preliminary report on the inheritance of the duration of the vegetation period is a small part of the material we obtained during the last period of our work.

We give the data of characteristic crosses studied under strictly identical experimental conditions on the plot of the Institute at Krasnodar.

Crosses given in Table 1 show three types of inheritance of the length of the vegetation period measured by the number of days from the transplanting of the plants into the field until the appearance of the first flower:

1) The vegetation period of the first generation is shorter than that of the early parent.

2) The vegetation period of the first generation is approximately equal to that of the early parent.

3) The vegetation period of the first generation is of an intermediate character (it is shorter than the vegetation period of the late parent and longer than that of the early one).

Among the numerous crosses, cases of the first and the second type of inheritance were the more frequent.

Below we give a detailed table of data on the intermediate type of inheritance (Table 2).

In the second generation we observe an extraordinarily wide range of variations in the duration of the vegetation period. This is shown in Table 3.

In the cross shown in Table 3 the first generation is an intermediate type of inheritance of the duration of the vegetation period, while in the second generation the plants become earlier. Moreover, a clearly expressed transgression of this character is to be observed here, a phenomenon occurring in cases of the inheritance of other quantitative characters: height of plant, number of leaves, dimension of leaf, etc. The phenomenon of the transgression of characters is of great importance in the plant breeding work.

Table 1

Inheritance of Duration of Vegetation Period in F_1-Generation of Hybrids

Nos	Crosses	Number of days before flowering
1	Varatik 26 *(P)*	43.3
2	Varatik 26×Giant 687 F_1	40.3
3	Giant 686 *(P)*	Did not bloom before frost. Over 100 days
4	Atropurpurea 1 200 *(P)*	66.8
5	Atropurpurea 1 200×Brazil 1594 *(F_1)*	61.2
6	Brazil 1594 *(P)*	52.6
7	Brazil 1594 *(P)*	52.6
8	Brazil 1594×Macrophylla 506 *(F_1)*	47.9
9	Macrophylla 506 *(P)*	48.4
10	Macrophylla 506 *(P)*	48.4
11	Macrophylla 506×Virginia dark 1631 *(F_1)*	50.0
12	Virginia dark 1631 *(P)*	82.5
13	Virginia dark 1631 *(P)*	82.5
14	Virginia dark 1631×Kuban 1148 *(F_1)*	62.2
15	Kuban 1148 *(P)*	63.3
16	Kuban 1148 *(P)*	63.3
17	Brazil 1594×Kuban 1148 *(F_1)*	54.4
18	Brazil 1594 *(P)*	53.4
19	Brazil 1594 *(P)*	53.4
20	Brazil 1594×Virginia dark 1631 *(F_1)*	58.4
21	Virginia dark 1631 *(P)*	95.5
22	Virginia dark 1631 *(P)*	95.5
23	Atropurpurea 1 200×Virginia dark 1631 *(F_1)*	76.6
24	Atropurpurea 1200 *(P)*	66.8
25	Atropurpurea 1200 *(P)*	66.8
26	Atropurpurea 1200×Havanna 1708 *(F_1)*	61.73
27	Havanna 1708 *(P)*	63.44
28	Kuban 1148 *(P)*	63.3
29	Macrophylla 506×Kuban 1148 *(F_1)*	49.30
30	Macrophylla 506 *(P)*	50.9
31	Brazil 1605 *(P)*	74.4
32	Brazil 1605×Virginia bright 1461 *(F_1)*	74.24
33	Virginia bright 1461 *(P)*	89.9
34	Virginia bright 1461 *(P)*	89.9
35	Virginia bright×Macrophylla 506 *(F_1)*	54.40
36	Macrophylla 506 *(P)*	49.2

Transgressions in the duration of the vegetation period in the second and further generations were to be observed in all the other crosses investigated when a great number of plants was used for the experiment.

The nature of the inheritance of the duration of the vegetation period is still more clearly manifested in the second and third generations of the cross between the Crimean variety of Dubec tobacco and Khinensis, subjected by us to detailed genetic investigation in 1935. The results are shown in Table 4.

In this cross, as well as in all the foregoing ones, the plants of the parent forms of the F_1, F_2 and F_3- generations were sown and transplanted into the field simultaneously. The third generation is represented by the posterity of many plants showing the whole variation range of the duration of the vegetation period in the second generation. The table shows also the data

Table 2

Inheritance of Duration of Vegetation Period in F_1-Generation

	Crosses \ Number of days before flowering	31	32	34	36	38	40	42	44	46	48	50	52	54	56	58	60	62	64	66	68	70	72	74	76	78	80	82	84	86	88	90	92	94	96	Average of days before flowering $M \pm m$
1	Giant 687 *(P)*	This parental form did not flower during the whole summer, i. e. more than 100 days																																		
2	(Giant 687 × × Shortstemmed 1201) *(F_1)* . .	—	—	—	—	—	—	—	2	3	5	7	8	5	4	—	—	—	4	—	1	1	1	2	2	—	—	—	—	—	—	—	—	—	—	56.9 ± 1.33
3	(Shortstemmed 1201× × Giant 687) *(F_1)*	—	—	—	—	—	—	—	—	5	8	8	2	7	7	2	2	1	—	1	1	—	2	2	1	—	—	—	—	—	—	—	—	—	—	56.2 ± 1.01
4	Shortstemmed 1201 *(P)*	—	2	—	2	5	6	10	5	2	3	4	—	1	—	—	—	—	1	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	42.5 ± 0.90
5	Shortstemmed 1201 *(P)*	—	2	—	2	5	6	10	5	2	3	4	—	1	—	—	—	—	1	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	42.5 ± 0.90
6	(Shortstemmed 1201 × × Samsun 27) *(F_1)*	—	—	—	—	—	—	3	5	5	4	6	5	3	2	3	2	3	1	1	1	2	1	—	1	—	—	—	—	—	—	—	—	—	—	53.0 ± 1.24
7	(Samsun 27 × × Shortstemmed 1201) *(F_1)* . .	—	—	—	—	—	—	3	1	8	8	12	7	2	6	—	2	1	1	—	—	—	—	—	1	—	—	—	—	—	—	—	—	—	—	59.3 ± 0.81
8	Samsun 27 *(P)*	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	5	6	4	5	1	3	3	5	1	2	—	1	—	1	—	3	—	74.3 ± 1.37
9	Trapezund 709 *(P)*	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	1	—	1	1	2	2	1	2	2	—	8	1	4	—	2	1	83.7 ± 1.43
10	(Trapezund 709× × Shortstemmed 1201) *(F_1)* . .	—	—	—	—	—	—	1	5	4	10	10	3	4	1	3	1	2	3	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	51.6 ± 0.84
11	(Shortstemmed 1201× × Trapezund 709) *(F_1)* . . .	—	—	—	—	—	—	3	2	6	9	9	7	5	2	7	—	—	—	—	—	1	1	—	—	—	—	—	—	—	—	—	—	—	—	51.6 ± 0.74
12	Shortstemmed 1201 *(P)*	2	—	—	1	3	6	6	4	9	2	7	2	1	—	1	3	—	1	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	46.3 ± 1.03

Inheritance of Duration of Vegetation

Crosses \ Number of days before flowering	19—23	24—28	29—33	34—38	39—43	44—48	49—53
Shortstemmed 1201 *(P)*	—	—	—	4	3	36	20
(Shortstemmed 1201 × Dubek 44) *(F_1)*	—	—	—	—	—	7	22
Dubek 44 *(P)*	—	—	—	—	—	—	—
(Shortstemmed 1201 × Dubek 44) *(F_2)*	15	7	20	47	234	287	187

Inheritance of Duration of Vegetation

Nos	Crosses \ Number of days before flowering	32	33	34	35	36	37	38	39	40	41	42	43	44	45	46	47
1	Dubek 44 *(P)*	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
2	(Dubek 44 × × Khinensis 346) *(F_1)*	—	—	—	—	—	—	—	—	—	—	5	1	1	3	4	2
3	Khinesis 346 *(P)* . . .	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	1
4	(Dubek 44 × × Khinensis 346) *(F_2)*	1	—	2	—	1	—	1	2	1	—	—	2	2	3	5	8
5	(Dubek 44 × × Khinensis 346) *(F_3)*	1	6	5	2	8	1	1	1	8	4	6	4	11	6	8	18
6	(Dubek 44 × × Khinensis 346) *(F_3)*	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
7	(Dubek 44 × × Khinensis 346) *(F_3)*	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—

Table 3

Period in F_1 and F_2 Generations

54—58	59—63	64—68	69—73	74—78	79—83	84—88	89—93	Total of plants	Average of days before flowering $M \pm m$
18	2	4	2	—	—	—	—	89	50.3 ± 0.74
15	2	3	—	—	—	—	—	49	53.1 ± 0.71
—	4	30	18	7	10	7	—	76	71.7 ± 0.82
125	52	26	33	12	15	5	2	1067	4.79 ± 0.34

Table 4

Period in F_1, F_2 and F_3-Generations

48	49	50	51	52	53	54	55	56	57	58	59	60	61	62	63	64	65	66	67	Did not flower during 67 days	Number of plants in the experiment
—	—	—	—	—	—	—	—	—	2	2	2	4	1	5	10	—	—	—	—	91	117
—	1	4	1	6	1	9	3	16	4	5	3	16	3	2	3	—	—	—	—	45	138
—	—	1	—	3	1	4	5	—	6	1	10	—	6	—	8	—	—	—	—	55	101
4	2	4	4	4	4	24	11	13	17	31	11	15	16	12	9	8	—	—	—	268	485
8	2	8	5	11	12	18	19	7	25	15	9	8	26	3	6	17	—	—	1	93	383
—	—	—	—	—	—	—	—	—	—	—	—	1	—	—	1	—	—	—	—	43	45
—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	128	128

regarding the third generation which was obtained only from three plants of the second generation representing the most characteristic cases of inheritance. (See Table 4).

The flowering in this cross was checked until the first of August of the present year, when this table was drawn up, and the number of the plants that had not flowered by this date is shown in the table.

In this cross the duration of the vegetation period of the first generation is shorter than that of the early parent. In the second generation, the same complicated segregation and transgressions as in the foregoing cross are to be observed. The third generation shows more cases of transgression than the second.

We observe an analogous type of inheritance of the duration of the vegetation period in the other crosses studied during this year.

Thus, on the basis of the investigation of the material obtained during a number of years in studying the crosses of the most varied parental couples, we have come to the following quite definite conclusions on the inheritance of the duration of the vegetation period in tobacco:

1) The first generataion is characterized by a) a prevalence of early flowering; b) an intermediate type of inheritance and c) cases of the duration of the vegetation period being shorter than that of the early parent.

2) In the second and third generations there arise plants ripening earlier as well as plants ripening later than the original forms and the first generation.

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GEOLOGY

ON THE STRATIGRAPHY OF THE QUATERNARY DEPOSITS OF THE SOUTH-WESTERN PORTION OF THE ICE LOBE IN THE DON RIVER BASIN

By V. I. KROKOS

(Communicated by A. A. Borissiak, Member of the Academy, 29. IX. 1935)

The study of the quaternary system in the Ukrainian SSR made it possible to subdivide this system into a number of individual stratigraphical horizons which may be traced within vast areas. As far back as 1927, I revealed with the aid of boring a three-stage series of loess with two horizons of fossil soils (1) at the bottom of a pit on the plateau of the former Starobelsk district (the eastern outskirts of the Kharkov province). This series is to be classified as belonging to the Bug, Dnieper and Tiligul stages, while the fossil soils seem to belong to the Dnieper-Bug and the Tiligul-Dnieper intervals (2).

In 1935 P. K. Zamory made out the presence of four, possibly of five stages of loess by boring on the plateau in the same region (3).

Thus, the quaternary stage series of the Ukrainian SSR may be traced as far as its eastern boundaries.

The present note contains some information on the stratigraphy of the quaternary series of the south-western portion of the ice-lobe of the Don river basin in the area of the Voronezh province, contiguous to the Kharkov province.

The area under investigation borders upon the right bank of the Don river, South of the railway junction Liski, in the space enclosed by the Tikhaya Sosna and the Kalitva rivers, in the North-West and in the South respectively. It comprises both the glaciation zone and the adjacent, towards South-West, extra-glacial one.

The structure of the plateau of the glacial region was made clear by borings. The borehole on the plateau near the village Shcherbakovo (22 kilometers to the S.S.W. from Liski) exposed a three-stage loamy super-morainic series and a moraine along which tertiary sands were to be observed. The first stage, from the top downwards, is 3.65 m thick. It is a heavy carbonaceous buffish loam with pipes of lime. The second stage consists of carbonaceous loam with lime pipes; its thickness is 2.25 m. The third stage consists of pale brown loam with pipes of lime; its thickness amounts to 2.05 m. It gradually passes into the moraine. The latter, which is 3.55 m thick, consists of two horizons,—an upper one with boulders of crystalline

rocks, and a lower one, containing an abundance of small boulders of chalk and chalky marl*.

As outlined above, the moraine is underlain by tertiary sands.

The three stages of super-morainic loam may be parallelled with the three-stage super-morainic loess series of the Ukrainian SSR, viz. with the Bug and the Udai stages and the upper part of the Dnieper stage (²).

The stages of carbonaceous loams are separated from each other by two horizons of fossil soils of chernozem type, which shows that steppe conditions are characteristic for the epochs of their formation. They may be parallelled with the first and the second fossil soils of the super-morainic loess series of the Ukrainian SSR. Their age is to be determined as corresponding to the Udai-Bug and the Dnieper-Udai intervals.

It should be noted that in some profiles the loam of the Udai stage and the fossil soil formed thereon are deficient.

In the outcrop of Krivaya Poliana village 45 km S. W. from Liski light gray sand with quartz and sandstone pebbles, scarce and small, was found under the moraine; this sand is 0.8 m thick and seems to represent submorainic fluvioglacial deposits.

Thus, the glacial region is characterized by a weak development of fluvioglacial deposits.

So far, it has been impossible to single out the equivalents of the submorainic stage-like loess series of the Ukrainian SSR in the moraine area.

The structure of the quaternary series of the extraglacial region has been made clear by a borehole on the plateau near the village Lutsenkovo (25 km S.W. from the village Krivaia Poliana). At this point four stages of dark-buff and pale-brown carbonaceous loams were discovered. These have frequently pipes of lime and are separated by three horizons of fossil chernozem soils. The first stage is up to 3.85 thick. The thickness of the second and the third stages equals 0.45 m respectively. In the fourth stage 2.45 m were exposed to view. The above stages are to be parallelled with the Bug, Udai, Dnieper and Tiligul stages of the Ukrainian loess series. Attention should be paid to the low thickness of the second stage, and to the high thickness of the third stage, which fact is also characteristic of the Ukrainian SSR. The thickness of the first stage is very close to that of the Bug loess stage of the Ukrainian SSR. The three chernozem horizons which separate these stages are related to the Udai-Bug, Dnieper-Udai and Tiligul-Dnieper intervals respectively.

The deficiency of the second loam stage (the Udai stage) was established also with regard to the extraglacial region. Thus, on the plateau near the village Kopanki (12 km S.W. from the village Olkhovatka on the Kalitva river) three stages of carbonaceous loams were exposed to view by boring. They were referred to the Bug, Dnieper and Tiligul stages respectively.

The first (Bug) stage of the extra-glacial region also consists of heavy loam. In a borehole on the plateau N. E. from the village Kopanki, this stage consists of brownish olive-yellow heavy carbonaceous loam, prismatically cuneiform in structure, containing scarce lime pipes. The rock cleaves into thick horizontal plates.

On the slopes, deluvial loess-like loams with sharp-edged débris of Upper Cretaceous rocks and red-brownish loams, as well as weathering products of chalk marls were observed to occur.

Along the Kalitva river three flood-plain terraces were ascertained (in

* The area under investigation is situated in the region of the development of Upper Cretaceous deposits.

<table>
<tr><th colspan="2">Ukrainian SSR</th><th colspan="2">Voronezh province
S. W. part (Don river basin)</th></tr>
<tr><th>Glacial region</th><th>Extra-glacial region</th><th>Glacial region</th><th>Extra-glacial region</th></tr>
<tr><td colspan="2">Bug loess stage</td><td colspan="2">Heavy loam of the Bug stage</td></tr>
<tr><td colspan="4">Udai-Bug interval (fossil soil)</td></tr>
<tr><td colspan="2">Udai loess stage</td><td colspan="2">Loam of the Udai stage</td></tr>
<tr><td colspan="4">Dnieper-Udai interval (fossil soil)</td></tr>
<tr><td>Super-morainic portion of the Dnieper loess Stage</td><td rowspan="3">Dnieper loess stage</td><td>Loam equivalent to the super-morainic portion of the Dnieper loess stage</td><td rowspan="3">Loam of the Dnieper (Dnieper-Don) stage</td></tr>
<tr><td>Moraine of the Dnieper glaciation</td><td>Moraine of the Don glaciation</td></tr>
<tr><td>Sub-morainic portion of the Dnieper loess stage</td><td>?</td></tr>
<tr><td>?</td><td>Orel-Dnieper interval (fossil soil)</td><td rowspan="4">Not established</td><td>Tiligul-Dnieper interval (fossil soil)</td></tr>
<tr><td>?</td><td>Orel loess stage</td><td rowspan="3"></td></tr>
<tr><td colspan="2">Tiligul-Orel interval (fossil soil)</td></tr>
<tr><td colspan="2">Tiligul loess stage</td><td>Loam of the Tiligul stage</td></tr>
</table>

the region of the village Sheliakino). The lower flood-plain terrace consists of alluvial sands; it corresponds to the loess-less terrace of the Ukrainian SSR.

The second flood-plain terrace is covered by loess-like carbonaceous loam, whose thickness approaches 3 m and which is underlain by pale-grayish and dark yellow fresh-water loams. This terrace corresponds to the terrace with one loess stage in the Ukrainian SSR.

The third flood-plain terrace was met with in the shape of a small fragment in the region of the soviet farm Olkhovatskaya Sosna. It consists of two stages of loess-like loams separated by a horizon of fossil chernozem soil. This terrace is to be parallelled with the one covered with two stages of loess in the Ukrainian SSR.

The above profiles testify to the fact that the moraine of the Don glaciation is covered by a three-stage loamy series. An analogous state of things was long ago demonstrated in the territory of the Dnieper ice lobe, where the moraine is overlain by a three-stage loess series. This fact shows that the Don and the Dnieper ice lobes are of the same age.

Thus, the stage-like quaternary series of the Ukrainian SSR may be parallelled in the territory of the Voronezh province. The problem of a more detailed subdivision of the quaternary series in the Voronezh province and the genesis of its components may be solved in the course of further investigations.

The relationships of the quaternary series of the Voronezh province and the Ukrainian SSR are shown in the above table.

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